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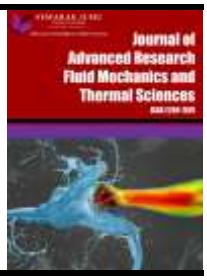

Towards Sustainable Food Packaging: A Review of Thermoplastic Starch (TPS) as a Promising Bioplastic Material, its Limitations, and Improvement Strategies with Bio-fillers and Essential Oils

Saiful Bahri Mohd Diah[1], Sharifah Hanis Yasmin Sayid Abdullah[1,2,*], Yew Been Seok[1], Ireana Yusra Abdul Fatah[1], Nor Iza A. Rahman[3], Fadli Hafizulhaq[4,5], Nur Nazira Alias[1]

[1] Faculty of Innovative Design and Technology, University Sultan Zainal Abidin, Kampus Gong Badak, 21300 Kuala Nerus, Terengganu, Malaysia
[2] Biomaterials and Nanomaterials Research Group, Smart Technology Research Cluster, University Sultan Zainal Abidin, Kampus Gong Badak, 21300 Kuala Nerus, Terengganu, Malaysia
[3] Faculty of Medicine, University Sultan Zainal Abidin, Kampus Kota, 20400 Kuala Terengganu, Terengganu, Malaysia
[4] Department of Agricultural and Biosystem Engineering, Universitas Andalas, Padang, Indonesia
[5] Research Collaboration Center for Nanocelulose between BRIN and Universitas Andalas, Padang, Indonesia

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ABSTRACT

Packaging industry is the dominant user of plastic that contribute highest amount of plastic waste entering our environment. Hence, alternatives such as bio-based plastic have emerged and becoming more commercialized. Thermoplastic starch (TPS) is one of the raw materials used in the production of bioplastic film. However, the major drawback of using TPS is due to its low mechanical, poor barrier properties, and brittleness. This review article provides summary of TPS as a choice of food packaging materials. It reviews recent studies on the improvements of TPS by incorporation of bio-filler and essentials oils. It also describes the impact on the TPS reinforced biofilm on film properties including mechanical, barrier and antimicrobial properties. This paper also discusses the performance TPS reinforced biofilm in ensuring shelf stability and perishability of food product for food packaging application. Finally, it also highlighted the challenge and opportunities TPS reinforced biofilm for the food packaging industry.

1. Introduction

Food packaging generally serves the four major purposes of containment, protection, convenience, and communication; yet each package is fitted to the needs of a particular food product [1]. The primary purpose of packaging is to shield its contents from external physical and chemical environmental factors like moisture, gases, odors, microorganisms, dust, impacts, vibrations, and pressure that may occur during transportation, handling, and storage, thereby prolonging the food's shelf life. This approach ensures efficient loading and transport throughout the entire supply chain while guaranteeing that consumers receive a secure, high-quality product. Moreover, effective

* *Corresponding author.*
E-mail address: shyasmin@unisza.edu.my

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packaging, often referred to as "silent salesmanship," offers an appealing and user-friendly product label to the end consumer [2].

Since its discovery, conventional food packaging made from plastics derived from fossil sources has been widely adopted to replace packaging made from materials like paper, glass, and metal due to its superior attributes, including versatility, mechanical strength, and cost-effectiveness [1,3]. Notably, various polymer types are utilized in food packaging, including polyethylene, polypropylene, polystyrene, polyester, nylon, polycarbonate, and vinyl polymers. These polymers offer both exceptional mechanical reliability and strength, making them crucial for food packaging applications and promoting their suitability for reuse and participation in waste-to-energy processes [4]. However, their accumulation can have adverse environmental effects and pose significant challenges. Common techniques for polymer disposal include landfill, incineration, and chemical treatment, all of which negatively impact the environment. Furthermore, their non-biodegradable nature poses a substantial threat to human health [5].

Hence, efforts have been continuously growing to explore the green alternative that could potentially replace the conventional food packaging material [6]. This has given rise to a burgeoning field of research in bioplastic production. Bioplastics are derived from renewable resources, are economically viable, abundant, readily accessible, and biodegradable. This paper reviews recent progress and improvements on reinforced starch-based bioplastic for food packaging applications. Their mechanical barrier properties and antimicrobial properties will also be discussed in the study.

2. Bioplastic

The European Bioplastics Organization (EBO) provides a definition for bioplastics, categorizing a material as such if it is either derived from renewable sources (bio-based), biodegradable, or exhibits both attributes [6]. The term "bio-based" pertains to materials or products that are wholly or partially sourced from renewable resources, specifically biomass [7]. Regarding to the origin resource, bio-based polymer can be classified into three main group which are biomass extract from polysaccharide resources (starch, chitosan, agar, and gum), polymer synthesised from biobased monomers through fermentation processes followed by polymerization of monomer product (e.g., polylactic acid), and polymers produced from micro-organism biochemical synthesis in microbial cells (e.g., polyhydroxylbutyrate) as shown in Figure 1 [8].

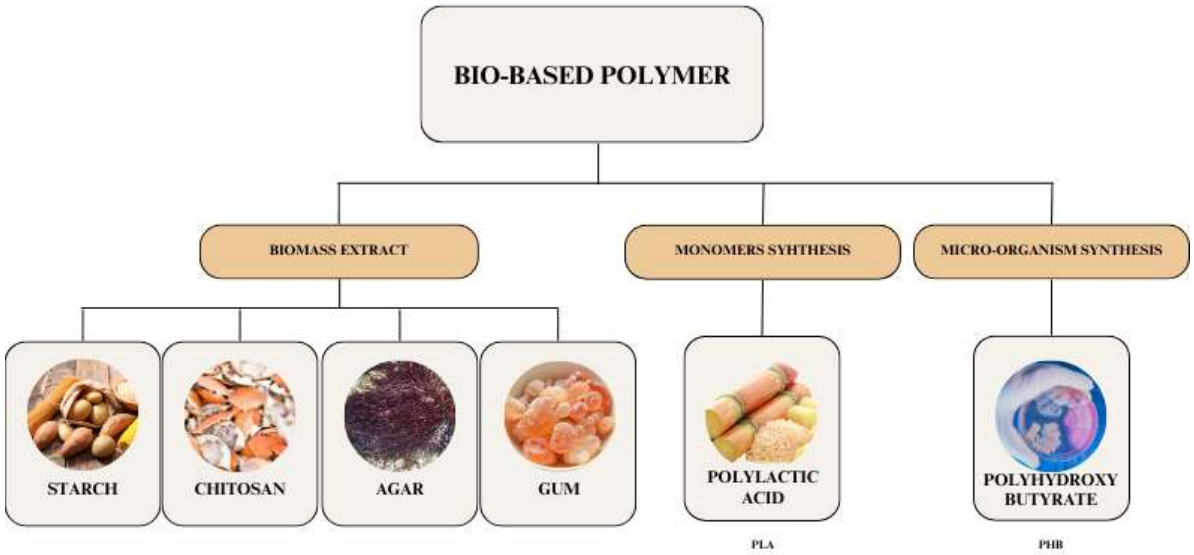


Fig. 1. Classification of bio-based polymers

2.1 Starch

Starch is a readily accessible and readily biodegradable natural resource that is not only renewable and sustainable but also abundant and cost-effective [9]. Starch primarily consists of two glucose macromolecules, namely amylose and amylopectin. However, functional and structural dissimilarities are present between different types of resources, consequently, the suitability of starch as a raw material for bioplastics hinges on its unique structure and composition [10,11]. In order to acquire thermoplastic starch (TPS), plasticizers such as water and glycerol, and sorbitol are added. TPS is mostly use as a constitute an alternative for polystyrene.

2.2 Chitosan

Chitosan is a functional biopolymer with intrinsic antimicrobial and antioxidant properties which enable its as high potential alternative material for biodegradable active food packaging [11]. Chitosan is derived from chitin by deacetylation via alkaline treatment method, composing N-acetyl-D-glucosamine (acetylated unit) and β-(1,4)-linked D-glucosamine (deacetylated unit) [12,13]. Similar to starch, chitin is among the most plentiful polysaccharides in nature, primarily sourced from biological waste like crustaceans and insect exoskeletons, the cuticles of certain arthropods, and the cell walls of fungi [14,15]. To enhance the mechanical attributes, water vapor permeability, antioxidant properties, and antibacterial efficacy in films designed for food packaging applications, chitosan can be functionalized via grafting or crosslinking [15].

2.3 Agar

Agar, derived from marine red algae like Gelidium and Gracilaria spp., is a promising polysaccharide for creating biodegradable antimicrobial packaging films [16]. Its chemical structure consists of two key components: agarose, a linear polysaccharide made up of repeating units of D-galactose and 3-6, anhydro-L-galactose, linked through alternating α-(1−3) and ß-(1−4) glycosidic bonds, and agaropectin, a slightly branched and sulphated polysaccharide [17]. Agar is valued in packaging film production for its robust mechanical properties and moderate water resistance. It is also commonly blended with other biopolymers such as starch, chitosan, and gelatin to improve the mechanical characteristics of these packaging materials.

2.4 Gum

Polysaccharide-based gums are derived from the endosperm of various plant seeds, primarily from the Leguminosae family. Most of these polysaccharide gums are categorized as galactomannans, which are composed mainly of the monosaccharide's mannose and galactose. Depending on the plant source, mannose components form linear chains that are linked to galactopyranosyl residues, forming side chains [18]. Gums are obtained from different parts of plants, including seed epidermis, leaves, and bark [19]. Gum Arabic, for instance, has a history dating back over 5000 years [20,21]. While gum-based edible packaging is recognized as an effective means to preserve the quality of fruits and vegetables, ongoing efforts persist to enhance the properties of these films [21].

2.5 Polylactic Acid

PLA, a biodegradable thermoplastic polyester, originates from eco-friendly, non-toxic, renewable sources. It is presently regarded as one of the most promising polymeric materials for practical use, serving as a viable substitute for low-density polyethylene (LDPE), high-density polyethylene (HDPE), polystyrene (PS), and polyethylene terephthalate (PET) [22]. Typically, PLA is manufactured by converting carbohydrates like corn or sugarcane, followed by a fermentation process to produce lactic acid. PLA pellets can be obtained through either the direct polycondensation of lactic acid monomers or through the ring-opening polymerization of lactide. Lactic acid (also known as 2-hydroxypropionic acid, or LA) is a chiral molecule with two optical isomers: L- and D-lactic acid. There are three distinct stereochemical compositions of lactide: L,L-lactide, D,D-lactide, and L,D-lactide [23]. The stereochemical composition significantly influences the ultimate properties of the polymer. The United States Food and Drug Administration (FDA) has classified PLA as "generally recognized as safe" (GRAS), making it a safe option for all food packaging applications [24].

2.6 Polyhydroxylbutyrate

PHB has been produced through the bacterial fermentation method under limited culture conditions using microorganism's activities of Alcaligenes, Azobacter, Bacillus, and Pseudomonas [25]. The properties of PHB properties are like that of the conventional petroleum-based polymers such as polypropylene (PP), polystyrene (PS), polyethene (PE) and polyethene terephthalate (PET) [26,27]. Apart from being biodegradable and non-toxic, PHB possesses highly crystalline nature that makes it more attractive for alternative plastic application [27]. PHB is used for several products including razors and bottles and biomedical items including sutures and bone implants [28]. However, little information and application can be found in Table 1 regarding its usage as food packaging due to its moderate barrier, thermal, and mechanical properties.

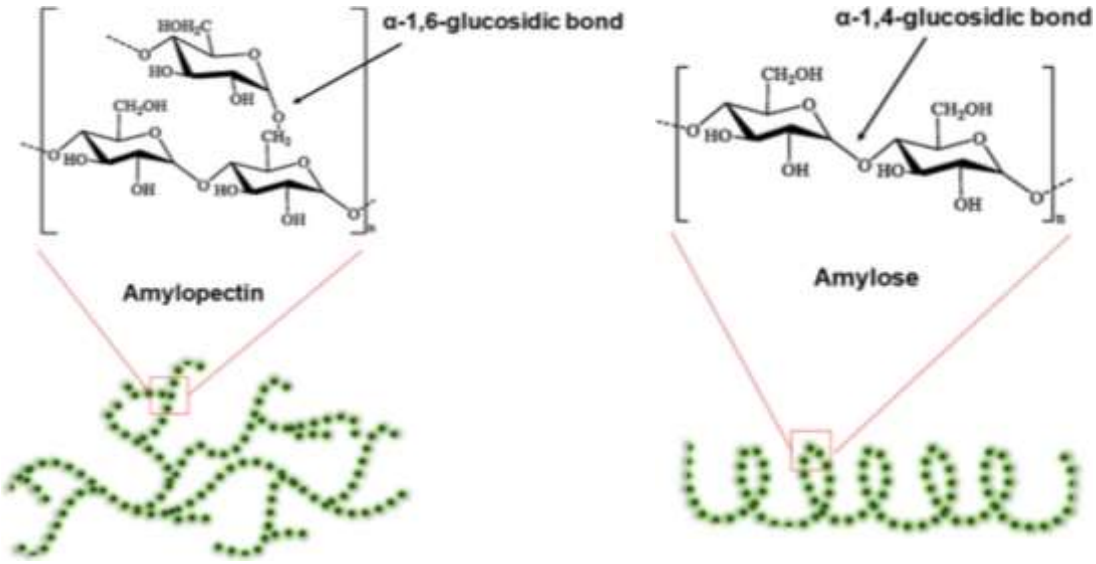
Table 1
Common applications for various types of bioplastics

Bioplastic	Source	Common application	Reference
Starch	Corn, pea, yam	Functional edible films, frozen meat and poultry wrapper, and bag.	Chang *et al.,* [29] Pająk *et al.,* [30] Dang and Yoksan [31]
Chitosan	Chitin (e.g., crustaceans, prawn, shrimp, crab, and etc.)	Antimicrobial films, edible film.	Mendes *et al.,* [32] Negm *et al.,* [33] Freitas *et al.,* [34]
PLA	Lactid acid that derived from (e.g., sugarcane, sugar beet, corn, and etc.)	Mulch films and bag, compost bas, vegetable and salad containers.	Garcia-Garcia *et al.,* [35] Suwanamornlert *et al.,* [36] Mazur and Kuciel [37]
PHB	Bacterial fermentation (e.g., *Bukholderia xenovorans, Alcaligenes, Azobacter*, and etc)	Biocomposite films, Agricultural and food packaging, Paper coating.	Kavitha *et al.,* [28] Sirohi *et al.,* [38] Yeo *et al.,* [39] Rodrigues *et al.,* [40]

3. Starch for Food Packaging

Starch has been widely used in various fields especially in the production of edible films and coatings for food packaging to improve the quality of the final product, for example corn starch is the most consolidated bioplastic alternative in food packages [40]. Starch is a natural bio-based polymer produced as reserve carbohydrates which form as discrete granules. It is the most common type of

carbohydrate found in many constituent parts of plants (seeds, fruits, and tubers) and the second-largest source of biomass generated on the land after cellulose [41,42]. Starch is composed of glucopyranose monomers (six-membered-ring glucose units) that are linked by α-1-4 and α-1-6 bonds. This arrangement classifies starch into linear polysaccharide amylose molecules and branched polysaccharide amylopectin molecules, as depicted in Figure 2 [15].

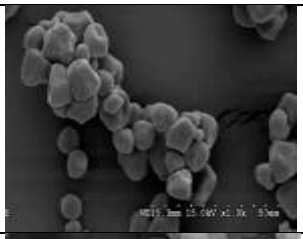
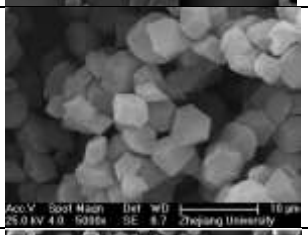
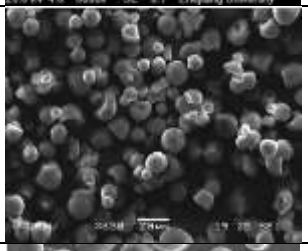
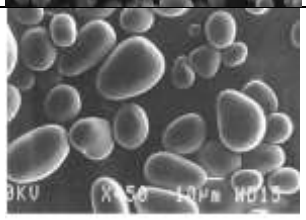


(a) amylopectin (b) amylose

Fig. 2. Schematic representation components of the starch [42]

Starch granules exhibit a semicrystalline nature and are composed of two primary components that are typically found together. The molecular weight of amylose can vary significantly depending on the source and the conditions of the extraction process, sometimes exceeding that of synthetic polymers by a factor of ten. As summarized in Table 2, amylopectin exhibits a notably higher molecular weight compared to amylose, typically accounting for only 15 to 30% [43-45]. Moreover, the specific properties, morphologies, and chemical structures of starch are influenced by the plant's origin and genetic makeup. Nonetheless, starch contains trace amounts of lipids, protein residues, and phosphorus.

Table 2
Properties of various types of starch

Source	Morphology	Properties		References
		Amylose (%)	Amylopectin (%)	
Corn		28.0	72.0	Marichelvam *et al.,* [46] Sun *et al.,* [47]
Rice		35.0	65.0	Sun *et al.,* [47]
Cassava		18.6	81.4	Marichelvam *et al.,* [46] Singh *et al.,* [48]
Potato		17.8	82.2	Singh *et al.,* [48] Purkan *et al.,* [49]

There are various methods of producing polymers from starch. In the synthesis of synthetic polymers, starch can be converted into compounds such as ethanol, acetone, or organic acids [15,49]. Moreover, it can be directly transformed toward a bioplastic monomer by hydrolysis or fermentation [50]. Although starch is a polymer that composed of continuous chains of amylose and amylopectin, yet it is not classified as a thermoplastic [51]. Hence, plasticizing process by addition of any plasticizer; water, glycerol, or sorbitol, then treated and sheared at high temperature (ranging from 140 to 160 °C) are compulsory to producing thermoplastic starch (TPS) and enable it to exhibit similar production ability as that conventional synthetic polymer [16,52,53]. Figure 3 illustrate the process of TPS development using solution casting method, solution casting is a prominent approach in lab-based TPS processing. It enables researchers to develop TPS films with precise composition and thickness control. This method is most appropriate for small-scale production and material testing in controlled conditions. Similar to that of synthetic thermoplastics, TPS is capable to be repeatedly softened and hardened into various shapes using heat and shear pressures. Despite being biodegradable, TPS has physical properties similar to traditional plastic used in food packaging applications in terms of odour and transparency [53].

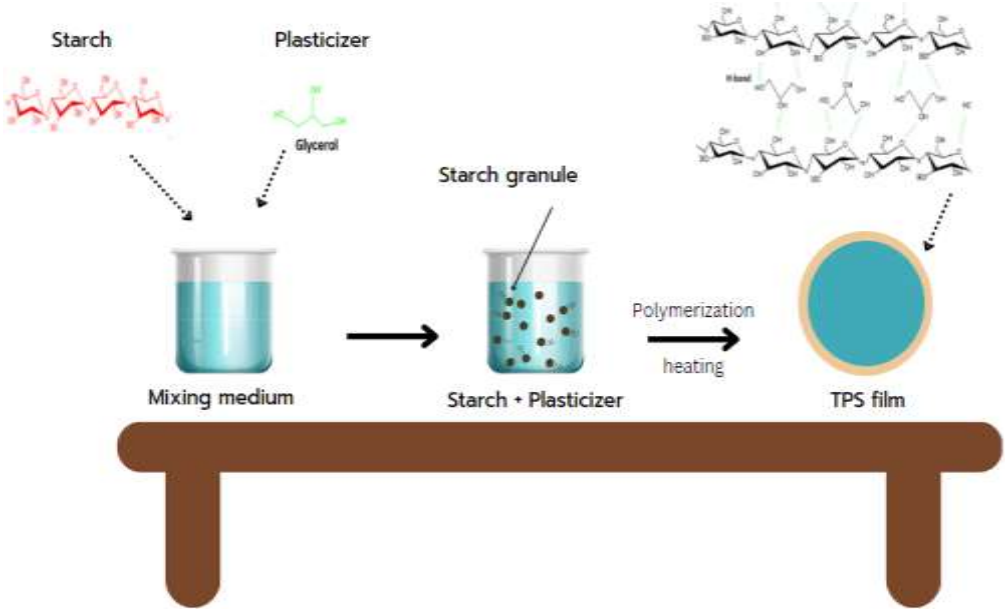


Fig. 3. Solution casting method for thermoplastic starch development

3.1 Limitation of Starch-based Packaging

Starch-based biodegradable products exhibit numerous disadvantages when compared to conventional thermoplastics, primarily due to the pronounced hydrophilic characteristics of starch [54]. Typically, starch chains are bound together by robust hydrogen bonds, rendering starch granules insoluble in cold water. However, the transition to TPS disrupts the crystalline structure of starch, allowing water molecules to interact with the hydroxyl groups, partially dissolving the starch granules [55]. This inherent hydrophilicity of starch results in diminished water stability and increased susceptibility to moisture, limiting the potential of starch-based materials.

TPS, owing to its molecular structure, demonstrates a relatively high glass transition temperature (Tg) and exhibits brittleness at ambient temperatures. With time, this brittleness increases due to retrogradation [55]. In addition, plasticizers such as glycerol have been shown to alter the crystallisation kinetics of starch and thus the final mechanical properties of TPS. The addition of plasticizers usually results in very ductile materials with relatively low ultimate tensile strength and modulus of elasticity [56]. Apart from elongation at break, the mechanical properties of materials based on TPS are quite poor compared to other thermoplastics, limiting their applicability to very narrow applications related to soft plastics [57]. Naturally, the processing conditions employed in material production also play a pivotal role in shaping its properties, as they can induce the degradation of starch molecules, for instance. Therefore, the ultimate mechanical properties of the material are heavily influenced by the storage duration prior to testing, as well as the concentration of plasticizers and the processing circumstances. As a result, numerous alterations have been studied in attempt to modify their features. The main methods for improving or reducing the poor qualities include the addition of reinforcements, coupling agent, chemical modification, and mixing with other polymers.

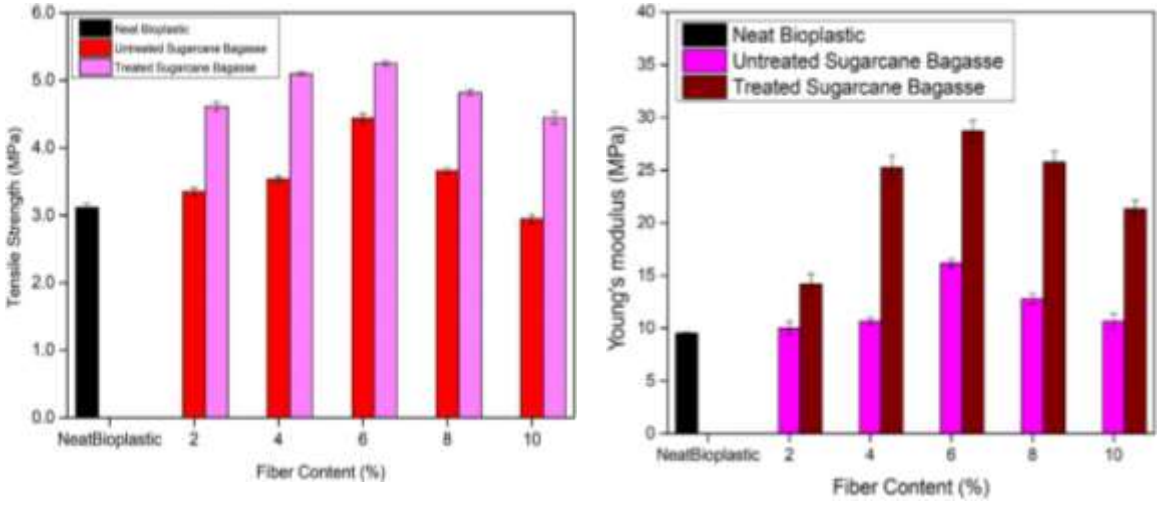
3.2 Natural Fiber Reinforcement

Biodegradable composites bonded with natural fibers offer a promising alternative to avoid synthetic fibres' toxins [58]. Over the last two decades, biodegradable composite research has grown incrementally as it reduces carbon footprint since these materials can be partially or completely

renewable and degradable [59]. One of the most extensively researched, reinforcements for improving thermoplastic starch qualities has been the insertion of natural fibres /organic lignocellulosic fibers [60]. Many lignocellulosic fibers have been assayed toward TPS matrix, including green coconut, sisal, hemp, cellulose fibers, nanofibers, and microfibers from recycled paper, and others.

Strengthening a starch matrix with natural fibers or lignocellulosic fibers has been proven to be highly effective. This is primarily due to the excellent compatibility between these fibers and the ability to form a 3D hydrogen bonding structure with starch components. As a result, biocomposite performance is significantly improved, leading to enhanced water permeability and mechanical properties, making them more suitable for use in food packaging materials [60,61]. Besides, natural fibers use less energy during growth and have adequate mechanical characteristics for a variety of applications [62]. In a study by Ochoa-Yepes *et al.,* [63] a thermoplastic film was created by incorporating lentil flour fibers using the film casting method. The composites that included lentil flour as a filler revealed that this fiber acts as a reinforcement for the starch-glycerol film, increasing its strength and protecting the food within. Menzel [64] reported the same results when developing starch-based films reinforced with cellulose fibers. The findings demonstrated that cellulose fibers significantly enhanced strength, with stress values reaching up to 1086 MPa. This phenomenon can be attributed to the interactions between starch and cellulose, which reduce the mobility of starch chains.

A study by Hamin *et al.,* [65] examines the "effect of chemical treatment on the structural, thermal, and mechanical properties of sugarcane bagasse as filler for starch-based bioplastic." The researchers measured the impact of chemical treatment on this fiber and found that the resulting thermoplastic material had a good improvement in tensile strength and young modulus. The results of the tensile strength and young modulus tests performed on various test samples are shown in Figure 4. The study discovered that the addition of natural fiber (sugarcane bagasse) enhanced the thermoplastic material's tensile strength and its elasticity. The ideal amount for this study, according to the researchers, was up to 6% sugarcane bagasse, they said. Researchers concluded that tensile strength decreased for percentages over 6% due to the failure of the fiber to establish an interlock bond with the film matrix.



(a) (b)

Fig. 4. (a) Tensile strength and (b) Young,s modulus of different loaded fiber [65]

3.3 Essential Oil as Starch-based Film Coupling Agent

Essential oils (EOs) are intricate mixtures composed of small, lipophilic chain compounds that are volatile obtained from plants through various extraction methods, including hydrodistillation, dry distillation, or mechanical extraction [66,67]. Essential oils are often referred to as etheric oils or volatility oils because they contain volatile organic compounds at room temperature [68]. The chemical characteristics of essential oils are challenging to standardize, as they vary depending on factors like the botanical sources, origin, production facility, extraction processes, and storage conditions [69]. In addition, parts of the extracted plant such as (whole fruits, flowers, bark, leaves, stems, pericarp, seed, or root) influence the composition of the essential oils [70,71]. Terpenes are the most predominant component in essential oils; terpenes are highly volatile secondary metabolites of plants produced by isoprene units. Terpenes become terpenoids after being biochemically changed [42]. However, essential oils do not solely consist of terpenes; they also contain numerous other elements, including phenols, aldehydes, ketones, carbohydrates, ethers, and alcohols, which play a pivotal role in conferring essential oils with their noteworthy biological activities [72]. Essential oils (EOs) are commonly employed for their antimicrobial properties, primarily attributed to the presence of hydrophilic functional groups and/or lipophilicity [73-76]. These attributes enable EO molecules to traverse bacterial membranes and access the interior of the cell, resulting in an inhibitory effect [77,78].

The phenolic constituents found in EOs also trigger an antimicrobial response against foodborne pathogens by disrupting the cell membrane and impeding cellular functions. Ultimately, this disruption leads to cell death as a consequence of intracellular substance leakage [42]. There has been a noticeable surge in the use of essential oils (EOs) as additives in the production of food packaging, with the objective of substituting synthetic additives [79]. For example, when EOs is included in packaging, the EOs contributes to antimicrobial and antioxidant activity through potential migration into the food over time. As a result, the shelf life of the food is extended, and their nutritional and sensory qualities are improved [80]. that the challenge of standardizing EOs results in packaging materials with diverse properties. Therefore, it is crucial to assess the impact of each type of essential oil on the mechanical, thermal, barrier, and structural characteristics of bioplastic. Numerous researchers have reported that the incorporation of EOs into biopolymer-based films can substantially modify the films' physicochemical properties and confer functional antimicrobial and antioxidant properties, all contributing to the enhancement of film performance. For instance, the addition of essential oils from cinnamon bark, clove, and oregano to blends of gelatin and cassava starch, plasticized with glycerol, resulted in improvements in water vapor resistance, oxygen barrier properties, and transparency, leading to films with superior functional attributes [81].

4. Properties of Starch-Based Polymer

Thermoplastic starch materials are used in the food industry for packaging respiring food (fruits, and vegetables) and non-respiring food (meats, fish, and cheese), Smaoui *et al.,* [82] suggesting that they have good prospects for commercial application. Different types of food require different film properties. To avoid undesirable metabolic changes, respiring foods (fruits, and vegetables) require packaging capable of maintaining oxygen levels while removing the form of water vapour. On the other hand, non-respiring foods (meats, fish, and cheese) need to prevent oxygen and water vapour permeability from entering the packages, to avoid microbial contaminations.

4.1 Mechanical Properties

The mechanical properties of thermoplastic starch film are one of the critical components in the manufacture of film packaging due to their beneficial effects in storage and trading. Therefore, to ensure the safety of food in food packaging applications, films must be strong, resilient, and ductile. Tensile strength (TS) and elongation at break (EB) are the key mechanical parameters of biopolymer-based films that are measured. TS stand for the film's resistance to tension forces, and EB assesses its ability for stretching, which denotes flexibility [77,83]. The study summarizes the impact of fibers reinforcement and essential oils coupling on the mechanical properties of the thermoplastic starch films. The researchers noted that the incorporation of essential oils led to structural modifications and changes in the film matrix's intra- and intermolecular bonds, resulting in significant alterations in the films' mechanical behavior [84]. The influence on TS and EB was highly contingent on the specific type of essential oil used, the film's composition, and the presence or absence of other formulation components. Table 3 below presents an overview of the effects of essential oil incorporation on film mechanical strength.

Table 3
Mechanical properties of starch-based film

Film Materials	Mechanical Properties		References
	Tensile Strength (MPa)	Elongation at Break (%)	
Cassava starch + Cinnamon Eos + Glycerol + Montmorillonite clay	2.32 ± 0.40 to 1.05 ± 0.16	264.03 ± 35.06 to 191.27 ± 22.62	Souza *et al.,* [85]
Corn wheat + Lemon oil + surfactant (Tween 80 and Span 80)	15.34 ± 1.40 to 11.18 ± 1.03	31.10 ± 0.37 to 36.30 ± 0.38	Song *et al.,* [86]
Potato starch + Lavender Eos + Furcellaran + Gelatin	70.2 ± 2.8 to 50.8 ± 2.8	20.1 ± 3.0 to 20.3 ± 2.0	Jamróz *et al.,* [87]
Pea starch + PVA + I. Oregano Eos II. Neem Eos	I. 26 ± 2 – 4.3 ± 0.13 II. 26 ± 2 – 7.2 ± 0.6	I. 40 ± 4 – 42 ± 11 II. 40 ± 4 – 44 ± 7	Cano *et al.,* [88]
Tapioca starch + Cinnamon Eos	0.69 ± 0.06 to 1.23 ± 0.09	159.91 ± 2.44 to 58.56 ± 1.23	Utami *et al.,* [89]
Corn/ Octenylsuccinated starch + Soybean	4.38 ± 0.07 to 3.58 ± 0.19	45.67 ± 11 to 17.42 ± 5.59	Gao *et al.,* [90]
Cassava starch + Cassava bagasse	4.70 ± 0.00 to 10.78 ± 0.00	-	Edhirej *et al.,* [91]
Corn starch + Corn husk	6.88 ± 0.00 to 12.84 ± 0.00	-	Ibrahim *et al.,* [92]
Arrowroot starch + Arrowroot bagasse	2.42 ± 0.00 to 15.22 ± 0.00	46.62 ± 0.00 to 6.21 ± 0.00	Tarique *et al.,* [93]
Corn starch + Nile rose fiber	3.83 ± 0.34 to 17.91 ± 0.56	9.60 ± 2.04 To 15.90 ± 3.50	Ibrahim *et al.,* [94]

Souza *et al.,* [85] investigated the effects of introducing cinnamon and clove essential oils into cassava starch films were examined, with the goal of developing active packaging. The analysis of the samples demonstrated that the addition of essential oils to the film reduced its tensile strength (TS) while increasing the elongation at break (EB) percentage when compared to the control film that did not contain essential oils. The control film's initial TS strength was 3.96 MPa, and its EB was 123.61%. The molecular makeup and mechanical characteristics of a corn/wheat starch-based biopolymer reinforced with lemon oil and surfactant for use in active packaging were studied by Song *et al.,* [86], these attributes included TS and EB of corn/wheat reinforced lemon EOs with Tween 80 and Span 80. From the study, it is found that increasing amount of lemon oil significantly reduced the tensile value, and in contras increasing the EB value. However, the incorporation of surfactant (Tween 80) to the film matrix reduced the EB value from 32.10 to 26.50 %. Another study investigated the influence of incorporating lavender essential oils into starch, furcellaran, and gelatin (S/F/G) films on their mechanical characteristics. With an increasing concentration of oil, the EB values remained constant, whereas the TS value saw a significant decrease to 50.8 ± 2.8 MPa. This suggests that a reduction in tensile strength could compromise the mechanical strength of the resulting biofilm [87].

A different investigation was conducted to evaluate the physical and antibacterial characteristics of starch-PVA mix film followed by the introduction of various concentration of Neem and Oregano EOs. EOs provide natural antibacterial properties and cutting-edge options for food packaging. The results showed that adding Neem and Oregano essential oils to the film matrix reduced the mechanical strength of the film. In comparison to the control film, the researchers observed a significant decrease in tensile strength, reaching up to 75% in films containing the highest concentrations of Neem and Oregano essential oils. On the other hand, the elongation at break (EB) percentage increased slightly for both types of essential oils at the lowest concentrations, but it began to decline as the essential oil concentration increased [88].

Researchers examined the mechanical characteristics of a tapioca film that contained cinnamon bark essential oil [89]. Only up to a 1% concentration of essential oil increase the TS (0.69 ± 0.06 to 1.57 ± 0.24) MPa before it began to decline as the concentration of essential oil rose (1.57 ± 0.24 to 1.23 ± 0.09) MPa. The TS of the film increased as the water content of the film changed. The introduction of essential oils into the film solution led to a reduction in the film's tensile strength, resulting in the formation of a heterogeneous film structure. As the essential oil concentration increased, the elongation at break (EB) also decreased, from 159.91 ± 2.44% to 58.56 ± 1.23%. This may be due to the development of a compact structure in the presence of essential oils within the polysaccharides, consequently reducing the EB value. Gao *et al.,* [90] observed a similar trend in the behavior of tensile strength (TS) in a film composed of corn/octenylsuccinated starch and incorporated soybean oil. The TS increased from 4.38 ± 0.07 MPa to 6.54 ± 0.25 MPa with a soybean oil concentration of up to 1%, before decreasing as the soybean oil concentration reached 2% (6.54 ± 0.07 MPa to 3.58 ± 0.19 MPa). The reduction in EB value was also noted as the soybean oil concentration increased, with the highest EB value recorded at 1.5%, which was 71.84 ± 6.31%. This was attributed to strong molecular interactions among starch molecules, resulting in a cross-linking effect that reduced the starch’s molecular mobility.

In contrast, the addition of fibers can have a contrasting effect compared to the addition of essential oils, notably in terms of enhancing the tensile strength of thermoplastic starch films. A noticeable increment in the tensile strength of cassava starch-based films was evident with an escalation in cassava fiber content from 3% to 6%. However, a decrease in tensile strength was observed at a 9% loading [91]. The highest tensile stress, reaching 10.78 MPa, was achieved with a 6% cassava fiber loading, surpassing the tensile strength of the control film, which measured 4.7 MPa. Conversely, elevating bagasse content from 6% to 9% led to films with reduced tensile stress and a

decline in Young's modulus. In another study by Ibrahim *et al.,* [92] the impact of corn husk loading (ranging from 2, 4, 6, and 8 wt.%) in corn starch-based thermoplastics was examined. The findings showed that the 8% concentration resulted in higher tensile strength and modulus values, measuring 12.84 MPa and 639.62 MPa, respectively, while the 2% concentration exhibited the lowest tensile strength and modulus.

Tarique *et al.,* [93] assessed the mechanical properties of Arrowroot starch-based films incorporating varying amounts of Arrowroot fiber. The results revealed a substantial increase in the tensile and tear strengths of TPAS/AF composites, rising from 4.77 to 15.22 MPa and 0.87 to 1.28 MPa, respectively, in comparison to the control TPAS films. However, there was a notable decrease in elongation, which reduced significantly from 25.57% to 6.21%. A similar decrease in elongation was observed by Ibrahim *et al.,* [94] in the case of corn starch-based films reinforced with Nile rose fiber. This reduction was attributed to the blocking effect of the Nile rose fiber, which constrained the film's ability to stretch. Interestingly, this reduction in elongation was accompanied by an improvement in tensile strength, with a maximum tensile strength of 18 MPa achieved with a 60% fiber loading. The review demonstrates that the mechanical properties of thermoplastic starch, such as TS and EB, cannot be easily standardised because they behave differently depending on the source and material reinforcement of the film, and significant changes in the percentages of EOs and/or filler used may influence the mechanical value stability. The value of TS and EB reported in this study is summarized in Table 3.

4.2 Barrier Properties

In contrary with metal and glass packaging, thermoplastics are permeable at different degree especially to the water vapour. To avoid food contamination from external environment and prolong the shelf life of food product, the study on barrier properties of the films has become increasingly important in recent years. Water vapour permeability (WVP) and water contents are crucial barrier factors in the quality of food packaging materials [95]. The study observes that, the incorporation of essential oils and filler reinforcement to the film matrix has significantly affected the barrier properties of the thermoplastic film. Table 4 summarize the barrier properties of starch film reinforced with various essential oil or/and filler.

Table 4
Barrier properties of various starch-based biofilm

Film Materials	Barrier Properties		References
	Water Vapor Permeability (g mm m^{-2} d^{-1} kPa^{-1})	Water Content (%)	
Cassava starch + glycerol + citric acid	-	20-26	Ounkaew *et al.,* [96]
Pea starch + PVA +			Cano *et al.,* [88]
I. Oregano Eos	4.6 ± 0.6 – 4.9 ± 0.5	4.4 ± 0.2 – 4.33 ± 0.13	
II. Neem Eos	3.7 ± 0.3 – 4.1 ± 0.4	4.5 ± 0.3 – 6.70 ± 0.13	
Corn wheat + lemon oil + surfactant (Tween 80 and Span 80)	3.68 ± 0.05 to 3.08 ± 0.05	23.30 ± 0.34 to 10.08 ± 0.27	Song *et al.,* [86]
Cassava starch + Cinnamon Eos + glycerol	3.61 ± 0.69 to 14.79 ± 2.76	-	Souza *et al.,* [85]
Potato starch + furcellaran + Lavender Eos	-	74.48 ± 5.04 to 68.81 ±6.44	Jamróz *et al.,* [87]
Cassava starch + Tea tree Eos	13.33 ± 1.16 to 10.46 ± 1.70	-	Silveira *et al.,* [97]
Tapioca starch + Cinnamon Eos	23.52 ± 0.45 to 21.38 ± 1.29	-	Utami *et al.,* [89]
Corn/ octenylsuccinared starch + soybean oil	2.93 ± 0.10 to 2.46 ± 0.04	-	Gao *et al.,* [90]

Previously Ounkaew *et al.,* [96] assessed four levels of PVA/starch films containing extracted wasted coffee powder, glycerol, citric acid, and ZnO. The studied found that glycerol increased the biodegradability and water resistance of PVA/starch films. Cano *et al.,* [88] performed the experiment by combining pea starch with polyvinyl alcohol (PVA) that had been fortified with oregano and neem essential oils. The S-PVA mix films had moisture levels of 4.40% and 4.33% for solutions containing 1% and 10%, respectively. After being processed for a week, films containing bioactive compounds had lower moisture levels. After a week of processing, the films containing the most neem oil reached balance, researcher also reported the incorporation of essential oils into the film did not significantly affect the water vapour permeability standard [98].

Song *et al.,* [86] conducted an analysis of the moisture content in various films composed of a mixture of maize starch and lemon essential oils (LEO). With higher LEO concentrations, the films' water content and WVP value reduced. This can be attributed to reduced interactions between starch and the two functional groups of LEOs, resulting in diminished interactions between polysaccharides and water, thus lowering the water content. Surfactants in the polymer films had an impact on how much water they contained. When Span 80 was added to the films, the water content was dramatically reduced. Hydrophobic surfactants, like Span, can lower the amount of water in films made of hydroxypropyl methylcellulose by inhibiting the adsorption of water vapour. This phenomenon in this investigation can be attributed to the film formulation's low Tween concentration [86].

For the cinnamon effect, Souza *et al.,* [85] conducted experiments with varies increment quantity of glycerol, emulsifier, and EOs, resulting in higher permeability values (ranging from 3.61 ± 0.69 to 14.79 ± 2.76) g mm m^{-2} d^{-1} kPa^{-1}. Consequently, increasing the glycerol content in the starch films manufactured in this study may lead to a reduction in the assessed barrier properties. A lepidic

component introduced into the film formulation can act as a barrier. Therefore, it was unexpected that cinnamon EOs would be the factor causing increased permeability values. Films containing cinnamon EOs displayed water vapor permeability (WVP) and oxygen permeability coefficients ranging from (9.78 ± 1.40 to 14.79 ± 2.76) g mm m^{-2} d^{-1} kPa^{-1} and from (27.50 ± 0.60 to 143.47 ± 8.30) × 10^9 cm^3 m^{-1} d^{-1} Pa, respectively. Jamróz *et al.,* [87] assessed the water content of potato starch films at LEO concentrations of 0%, 2%, 4%, and 6%. The addition of LEOs resulted in decreased water absorption in comparison to the control, with the film's water content ranging from 68.81% to 72.28%. The film with the highest oil content (6%) had a water absorption rate of 68.81%, while the control film's water content was 74.48%. This reduction resulted from the interactions among LEOs, potato starch, furcellaran, and gelatin functional groups, which limited interactions between polysaccharides, water, and proteins, leading to reduced water content.

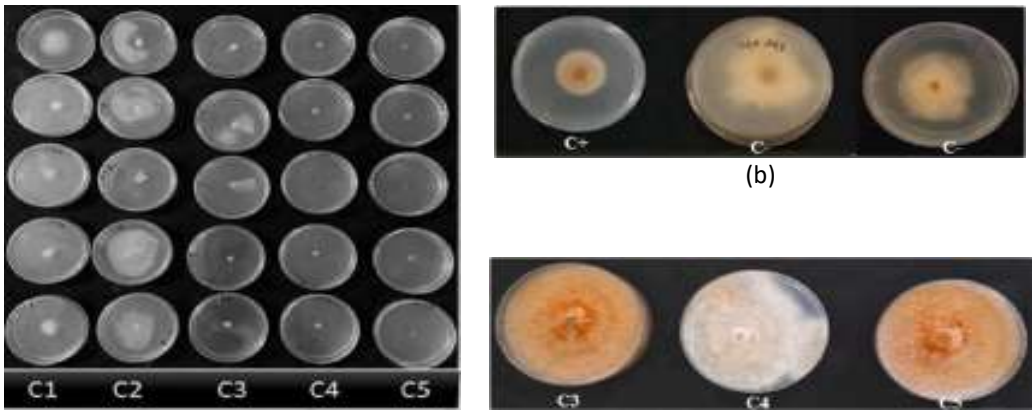
According to Silveira *et al.,* [97], the trend of the film's WVP was discovered to be reducing by the rise in tea tree essential oil (13.33 ± 1.16 to 10.46 ± 1.70) x10^{-11}g$Pa^{-1}s^{-1}m^{-1}$. The cohesiveness of the polymeric chain generated was shown by the WVP lowering. Because of the potent intermolecular interactions, the matrix's ability to diffuse water vapour was hindered. This reduction in WVP was attributed to the formation of a cohesive polymeric chain, impeding the matrix's ability to allow water vapor diffusion due to strong intermolecular interactions, according to Utami *et al.,* [89]. Lower WVP values are a criterion for better food moisture protection. Additionally, water vapour permeability only manifests itself under hydrophilic conditions. According to Gao *et al.,* [90], when the concentration of essential oil increased, the water vapour permeability (WVP) for the corn/octenylsuccinated composite film integrated with soybean oil somewhat reduced (2.93 ± 0.10 to 2.46 ± 0.04) x10^{-12}g.cm/cm^2.s. Pa. The film's hydrophobic nature, intensified by the addition of soybean oil, made it less prone to interacting with water, thereby reducing the WVP value. The hydrophilic phase of the film was disrupted by the inclusion of soybean oil, enhanced the diffusion path length and reducing the film's WVP. In summary, in most cases, an increase in the percentage of EO significantly reduced the water vapor permeability of the starch film.

4.3 Antimicrobial Properties

Another method for limiting, reducing, inhibiting, or killing food-spoiled or harmful microorganisms is anti-microbial packaging or also known as active packaging [99]. Active packaging requires biopolymers to have antimicrobial properties, and these properties can be improved by adding various natural substances such organic acids, curcumin, pectin, nano clays, and essential oils [42]. These organic substances have the capacity to inhibit lipophilic and functional characteristics by entering the cell membranes [100]. Anti-microbial compounds are often amalgamated into the polymer matrix to achieve anti-microbial action, which inhibits bacteria development [101]. Due to its numerous benefits, including improvements in food quality, shelf life, and food safety, this method has captured the attention of the food industry. Thus, extensive study has been conducted to assess the suppression of harmful microorganisms following the integration of the various EOs and reinforcement of natural filler in starch films.

Gonçalves *et al.,* [102] conducted both in vitro and in vivo experiments to assess the effectiveness of Melaleuca alternifolia essential oil in extending the shelf life of banana fruits. In the in vitro experiment, different concentrations of the essential oil were added to a Potato-Dextrose-Agar (PDA) medium inoculated with the pathogen C. musae. The results showed that all concentrations of the essential oil were more effective in controlling the pathogen than the commercial fungicide Tecto SC® and the control group without any essential oil. The essential oil-treated groups exhibited a significantly lower level of injury compared to the control group. Figure 5 shows the result obtained

with the MAEO spraying were superior to those obtained by the control (C-) and are still considered satisfactory when compared to the chemical control (C+) and can be considered an alternative in agroecological and/or organic crops.



(a) (b) (c)

Fig. 5. In vitro mycelial growth of C. musae under various M. alternifolia oil concentrations - Day 8 [102]

Cheng *et al.,* [103] recorded the efficiency antibacterial of yam starch-based incorporated with eugenol essential oil film against E coli, S. aureus, and L, monocytogenes bacteria on pork preservation. Figure 6 shows that there is presence of bacterial colonies at day 4 in control group, While the polysaccharides in the YD matrix encouraged the growth of bacteria, the group covered with YD film was more significantly spoilt than the control group. At day 7, the surface of the pork with YDE3 coating had minimal bacterial growth—just a few tiny colonies, distributed throughout which claimed to be extends the shelfs life of pork by more than 50%. In other study by Baek *et al.,* [104] a new antioxidant film was prepared using cowpea starch incorporated with different amount of maqui berry extract (0%-20%) and tested toward salmon sample within certain period time. The results of this study recorded that, retarded lipid oxidation of salmon samples was observed during storage time and the antioxidant activity increased with increasing content of maqui berry extract.

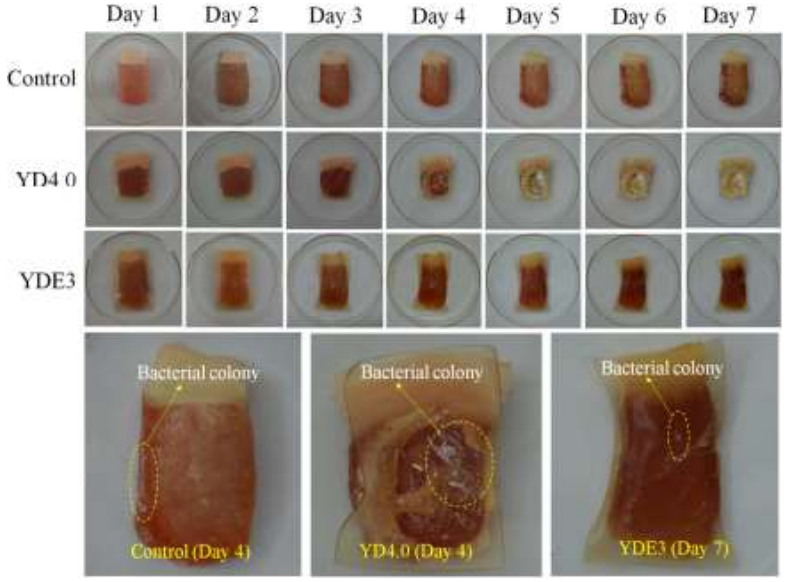


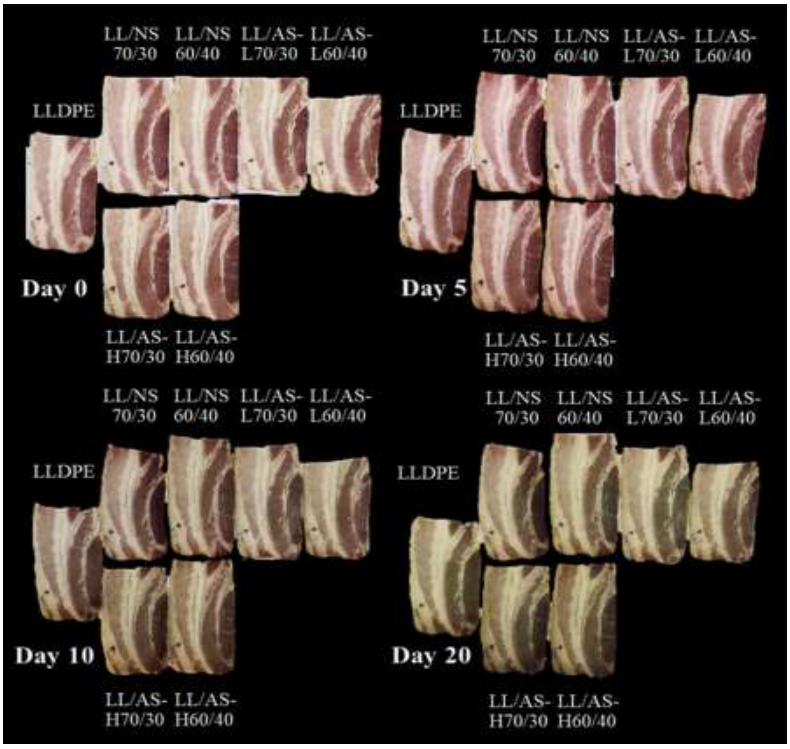
Fig. 6. Application of antibacterial film to pork preservation [103]

Additionally, in recent years essential was used as a natural antimicrobial substituting artificial preservative in biopolymer film. A vast of literature can be found concerning the evaluation of pathogenic microorganisms' inhibition activities with the incorporation of different essential oils in starch film. For example, the inhibition activities using essential oil against Salmonella enterica, Escherichia coli, Listeria monocytogenes, Pseudomonas aeruginosa, Enterobacter s. etc [34,61,84,105-107]. However, most of the applications of starch-based biodegradable material found in the literature have been carried out at laboratory scale rather than industrial scale. Finally, it is crucial to identify the type of essential oil used against the type of bacteria, whether Gram-positive or Gram-negative. Gram-positive bacteria are usually more sensitive to the inhibitory effect of essential oils, compared to Gram-negative bacteria.

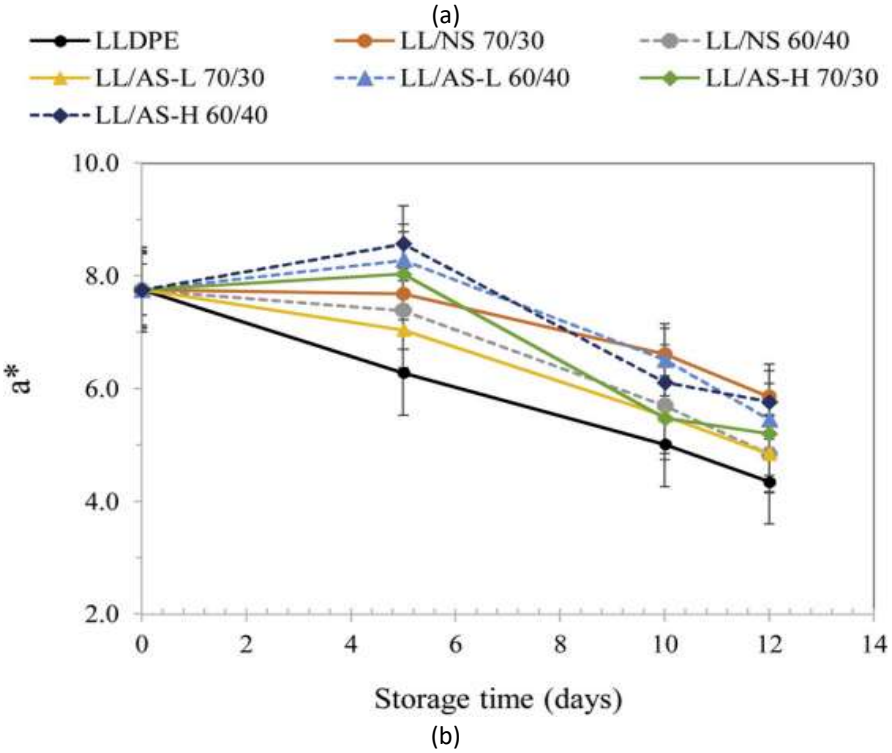
5. Storage Time and Conditions

An article reviewed by Bangar *et al.,* [60] compiled few remarkable works done by researchers, one of them as mentioned by Jabeen *et al.,* [108] stated that currently, non-perishable potato chips, biscuits, and pasta as well as perishable fresh fruits and vegetables are packaged with thermoplastics made of starch. Silva *et al.,* [109] stated that Fruit coatings made of cassava starch are widely used, but to improve their mechanical properties and water resistance, additional polymers were added to the films. According to López *et al.,* [110], in order to increase the shelf life and decrease microbial activity in strawberries that had been contaminated with the disease Botrytis cinerea, bio-composite films made of thermoplastic maize starch, MMT loaded, and EO were used. Biocomposite films containing carvacrol and thymol decreased microbial load 2.4 times, from an IC50 value of 14.16 (carvacrol) to 5.90 g/kg (carvacrol and thymol). For fresh meat products, thermoplastic maize starch and copper nanoparticle antimicrobial films have been an intriguing material.

Using the blown extrusion method, Panrong *et al.,* [111] created active films from acetylated cassava TPS and green tea for food applications. Prior to blown-film extrusion, different ratios of green tea and native acetylated TPS with varying degrees of substitution (DS) were co-extruded with LLDPE, specifically at ratios of 70:30 and 60:40. These films were then used to store soybean oil at 25 °C and 50% relative humidity for 28 days. Additionally, bacon slices were stored in these films at 4 °C for a period of 20 days. The results demonstrated that the green tea released from the starch matrices played a key role in preserving the color of the bacon, reducing microbial growth, and preventing the formation of metmyoglobin. Figure 7 provides visual representations of (a) the bacon's appearance and (b) its color changes during the 20-day storage period. The findings suggest that films with a higher TPS ratio were more effective in maintaining the redness of the meat. Films with an LL/AS ratio of 60/40 also exhibited a delay in meat discoloration and a more pronounced red color compared to other systems on day 20 (as illustrated in Figure 7(a). Notably, films with high DS LLDPE/acetylated starch showed the most remarkable ability to minimize lipid oxidation and release a higher quantity of phenols and antioxidants into the packaged soybean oil. Furthermore, these films enhanced the redness of the meat, delayed discoloration, and reduced the microbial load in sliced bacon by 1 log cfu/g. Films incorporating green tea also displayed reduced oxygen permeability, which inhibited the growth of aerobic bacteria and decreased the overall viable count in the meat.



(a)



(b)

Fig. 7. Changes in (a) appearance and (b) color (a*) of bacon during storage at 4 °C [111]

6. Challenge and Opportunity of Starch-Based Packaging

Plastics are popular in the food industry because of their low cost, durability, and flexibility. It is important to realise that the goal of producing thermoplastic starch films and using them in food is not to entirely supplant non-biodegradable plastics, as there are still many hurdles to assay. It is widely recognised that the low degradation rate of plastics and their subsequent accumulation in the

environment have triggered serious environmental, social, and economic consequences worldwide, as well as various debates on climate change, biodiversity loss and air and water pollution. Overall, the research has shown that EOs as active components improve the qualities of thermoplastic starch films used in food packaging. However, several issues remain to be addressed in selecting the appropriate biopolymer and production method for the film/coating, as well as in enabling their application on an industrial scale. Comprehensive studied are required to explore these issues to develop an effective method of film fabrication in industrial scale.

A noteworthy area for investigation pertains to the limited research on the long-term stability of essential oil components within a polymer matrix. For instance, how enduring is the antibacterial activity in such films? This is one of the questions that emerged during the review and will be a focus of future research. Furthermore, the standardization of essential oils used in film production has received inadequate attention, potentially affecting the reproducibility of the material. Additionally, there is a need to explore the effects of storage conditions, including temperature and relative humidity, on biopolymer-based packaging. Understanding migration and permeability to ambient gases and the roles of additives, fillers, and water vapor within the packaged food are equally crucial. Future research should consider these factors in food products or within food containers. Furthermore, dedicated research is essential to develop multifunctional antimicrobial engineered coatings/films with antiviral efficacy for evaluating their effectiveness against novel coronaviruses or related diseases.

7. Conclusion

Biodegradable polymers, whether sourced from natural biopolymers or synthetic bio-based polymers, are gaining traction in the realm of active food packaging. They serve to reduce the reliance on petroleum-based plastic packaging and meet the growing demand for fresh and health-conscious food products. Today's scholars and researchers prioritize ambitious goals, such as the reduction or elimination of plastic waste, as well as the establishment of an environmentally sustainable ecosystem. Notably, food packaging currently constitutes most of the plastic waste in our environment. Consequently, the adoption of biodegradable packaging in commercial markets becomes imperative in fostering sustainability and eco-friendliness. Beyond the environmental aspects, it is crucial to acknowledge that all food items, especially fruits and vegetables, face limited shelf lives, even when conventionally packaged in plastics. Considering the insights garnered from this review on food packaging applications, the introduction of essential oils into thermoplastic starch matrices brings forth a unique attribute, bolstering antimicrobial and antioxidant properties, which in turn offer significant advantages for food safety and preservation. Nevertheless, the persisting challenges related to compromised mechanical strength, water vapor resistance, and thermal stability present formidable obstacles for starch-based films seeking to establish a competitive foothold within the food packaging industry. However, this study has unveiled a promising solution in the form of reinforcing thermoplastic starch with natural fibers, effectively enhancing film properties and addressing these limitations. In summary, thermoplastic starch emerges as a promising contender in the field of food packaging, particularly in the realm of active food packaging.

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