

Biodegradable food packaging and film: a short review

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Abstract

Food packaging plays an important role in maintaining the quality of packaged food from any chemical, physical and environmental damage. Conventionally, plastic material is used in the food industry because of its large availability and excellent mechanical performance at low cost. However, the usage of plastic packaging in huge amounts creates environmental and health issues. The preparation methods of the biodegradable packaging or films have been reviewed. The properties of the packaging are varied depending on the ingredients of the films. The mechanical properties such as tensile strength of the biodegradable films can be improved by additive cellulose fibre or metal oxide composition. Metal oxide composition is added to the production of the film as reinforcement. The increase in metal oxide composition indicates the tendency to increase tensile strength. However, a decrease in elongation at break is observed as the percentage of modified cellulose increases. The addition of a natural bioactive compound such as antioxidant, antimicrobial and water vapour permeability also enhance the functionality of the film and provides intelligent communication with the consumer. The review provided information for readers to choose a suitable preparation method for biodegradable films. The article also offers data on materials selection based on the comparison of the film properties. Biodegradable packaging approach to mitigate the management of plastic waste which serves as an urgent crisis recently and supports in moving towards a sustainable environment.

1. Introduction

When it comes to food packaging, either raw, processed or cooked foods, various types of packaging are available. Food packaging must contain elements of protection, freshness and containment. The most common packaging materials used by food operators which contain these elements are plastic, aluminium and cardboard meanwhile glass may be a better choice for food that required high chemical stability and paper for food that is dry and to be consumed immediately.

The use of plastic packaging has increased in past decades. Many trading companies have invested in the industry of plastic packaging and thus there are different types of plastics available in the market now. Plastic bags are produced from the by-products of crude oil and they are recycled in many ways. However, the drastically increasing usage of crude oil causes some shortfalls. Furthermore, plastics are non-degradable materials, the decay of these materials takes up centuries and causes

pollution in the environment. The molecules in plastic material are constituted due to the strong bonding of intermolecular forces. This characteristic resulted in the structure of plastic neither decomposes nor corrodes for several thousand years. Other than food packaging, the plastic materials that are used in means of other purposes, such as in agriculture and the medical field contains harmful components. Improper disposal of plastic packages causes a lot of harmful and hazardous side effects, for instant water pollution which will endanger sea creatures, affect the water security for humans and animals that end up with the impact on ecology chain on the earth. Plastic pollution is emerging as a top threat to ocean ecosystems.

In order to overcome the side effects of plastic usages, researchers have started exploring their interest in materials that can replace non-degradable plastic. In 1888, biodegradable packaging materials made of cellulose, polylactide or starch blends have long been on

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the market (Mendes *et al.*, 2016). The benefits of biodegradable packaging include easily disposable, non-harmful toxins, biodegradable reduces carbon footprint, and reduce shipping cost. Green packaging not only reduces carbon footprint and environmental impact but is also beneficial after completing its purpose as packaging materials. In this case, biodegradable packaging reduces the shipping costs leading to less effort being expended (Mendes *et al.*, 2016).

This article reviewed the types of biodegradable films which are bio-based and petroleum-based plastic packaging in Section 2. Further discussion in Section 3 focuses on the properties of the biodegradable films including their structural properties, mechanical properties, antimicrobial activity and water absorption capacity. The benefits of additives in biodegradable food packaging and films are reviewed in Section 4 based on the types of preserved food.

2. Types of biodegradable plastic packaging

There are two types of biodegradable packaging which are bio-based plastic and petroleum-based plastics. Different types of plastic are made of different types of materials and have different applications. Table 1 summarizes the materials, application, advantages and disadvantages of those plastics.

The main difference between bio-based and petroleum-based plastics is the raw materials that are used to produce them. Petroleum-based plastics are polymers made from petrochemicals, whereas bio-based plastics are made from plant-based sources, like starch and cellulose. Petroleum-based plastics are more commonly known for being durable and long-lasting, though this is not an exclusive feature of this material. Table 2 summarizes petroleum-based plastics in terms of their contents, application, advantages and disadvantages.

There are many ways to produce polymer blends including solution blending, latex blending and melt blending. Solution blending is probably the most commonly used method for introducing alternative properties or functions to the core material. This method simply involves mixing two parts in a solution followed by electrospinning of which only one-part solution needs to be electrospinnable. Latex blends are the combination of capillary adhesion during the blending process with electrostatic adhesion in the final product. Latex blends are made using aqueous dispersions well within the “green chemistry” paradigm. Melt blending is the preferred method for preparing polymer nanocomposites of thermoplastics and the elastomeric polymeric matrix. Typically, the polymer is melted and combined with the

desired amount of the intercalated clay using an extruder (Harunsyah *et al.*, 2017).

Solvent-casting is a kind of century-old film-making process. For pharmaceutical applications, the active pharmaceutical ingredient (API) is either suspended or dissolved in a solution of polymers, plasticizers, and any other ingredients dissolved in a volatile solvent such as water or ethanol. In the casting method, a polymeric matrix is first solubilized in a relevant solvent with active components to form a solution to be cast onto the substrate. The solvent is then evaporated under controlled conditions to obtain an active film. The flow chart of the casting process is shown in Figure 1. Besides that, thermocompression is another film production method that is commonly used. Table 3 summarizes the common methods of film production by a few researchers.

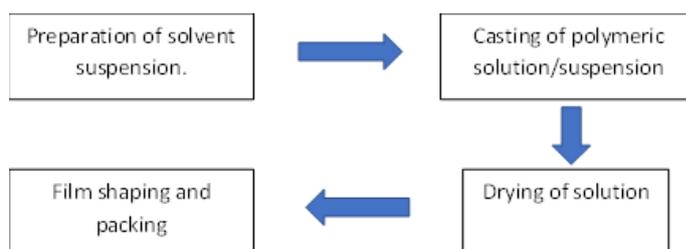


Figure 1. Flow chart of solvent casting method

3. Properties of biodegradable film/packaging

3.1 Structural properties

The common biodegradable materials for food packaging are gelatin, starch, chitosan, cellulose, and polylactic acid. The tensile strength, tear resistance, permeability, degradability, and solubility are some of the properties defining the selection and utilization of food packaging materials (Chisenga *et al.*, 2020). Generally, the original physical outlooks of food packaging or plastics are transparent, smooth and thin for flexibility in wrapping or sealing as in Figure 2(a). Pigment is added to lower the transparency. On the other hand, they are coated with an outer layer to provide an opaque outlook and printing of information about the food, i.e. name and brand of the packed food, ingredient and nutrition fact, manufacturer and instructions if any. The detailed morphology of the films can be observed using Transmission Electron Microscopy (TEM) or Scanning Electron Microscopy (SEM) as shown in Figure 2(b).

The crystallinity of the films is one of the factors to be concerned about as it will affect the dispersion of materials in the mixture or blend during the plasticization. The crystallinity and composition are detected using X-Ray Diffraction (XRD) and the crystal sizes can be calculated from the collected diffraction

Table 1. The types of bio-based plastic

Types	Content	Application	Advantages	Disadvantages	References
Polyhydroxyalkanoates (PHAs)	A class of biodegradable plastic naturally produced by various microorganisms	PHAs are applied in many fields, such as packaging, medicine, or agriculture.	bio-origin, biocompatibility, biodegradability	poor mechanical properties, high production cost.	Zulfiqar et al. (2017); Basnett et al. (2017)
Polylactic acid (PLA)	thermoplastic aliphatic polyester synthesized from renewable biomass, from fermented plant starch such as from corn, cassava, sugarcane.	It can be used as a food packaging polymer for short shelf-life products such as fruit and vegetables.	PLA is derived from a renewable resource	Improperly disposed PLA plastics can contaminate recycling processes	Hagen (2012); Melania et al. (2020)
Starch blends	thermoplastic polymers produced by blending starch with plasticizers	Used in food packaging, electronic devices, medical application.	Starch can replace petroleum-based polymers with natural ones.		Wang et al. (2007)
cellulose-based plastics	Cellulose bioplastics are mainly the cellulose esters, and their derivatives, including celluloid.	Major applications for cellulose plastics include thermoplastics, extruded films, eyeglass frames, electronics, sheets, rods	can improve mechanical properties, permeability to gas, and water resistance		Mostafa et al. (2015) Thakur (2014)
Lignin-based polymer composites	are bio-renewable natural aromatic polymers. Lignin is found as a by-product of polysaccharide extraction from plant material through the production of paper and ethanol	Applications to composites, including thermoplastic, thermosets, rubber, foams, bioplastics, nanocomposites, and lignin-based carbon fiber composites.	Inexpensive and renewable	Complex and disordered structure.	Faruk and Sain, (2015); Serma et al. (2019)

Table 2. The types of petroleum-based plastic

Types	Content	Application	Advantages	References
Polyglycolic acid (PGA)	A thermoplastic polymer and an aliphatic polyester.	Used in medical applications such as PGA sutures	degrade into its nontoxic monomer, glycolic acid, through hydrolysis.	Gorth and Webster (2011)
Polybutylene succinate (PBS)	A thermoplastic polymer resin	Used in packaging films for food and cosmetics. In the agricultural field, PBS is used as a biodegradable mulching film	A high heat resistance among the general biodegradability resin, and PBS has high compatibility with a fiber.	Rudnik (2013)
Polycaprolactone (PCL)	A synthetic polyester that is partially crystalline, having a low melting point	Used in production of specialty polyurethane	excellent mechanical properties, biocompatibility, bioactivity and faster degradation rate	Nair <i>et al.</i> (2017)
Poly(vinyl alcohol) (PVA, PVOH)	A water-soluble synthetic polymer	Used in food packaging, textiles coating, paper coating, and healthcare products.	excellent film forming, emulsifying and adhesive properties, resistant to oil and solvents, high tensile strength and flexibility, as well as high oxygen and aroma barrier properties.	Harekrishna <i>et al.</i> (2019)
Polybutylene adipate terephthalate (PBAT)	A biodegradable random copolymer, specifically a copolyester of adipic acid, 1,4-butanediol and terephthalic acid	Many products based on PBAT have been applied into many fields such as shopping bags, garbage bags, cutlery and mulch film	wide melting point, low elastic modulus and stiffness, but high flexibility and toughness.	Jiao <i>et al.</i> (2020)

Table 3. The casting method of biodegradable films.

Film production	Polymeric matrix	Temperature	Cooking time	Polymer ratio	References
Casting	Modified Cellulose Fibre-Reinforced with PVA	100°C	24 hours	PVA ratio from 5% - 95%	Sandeep <i>et al.</i> (2012)
	Bioplastic cassava starch film-based zinc oxide reinforced composite biopolymeric films	85 ± 5°C	15 mins Drying is carried out in oven at temperature of 60 °C for 5 hours	The content of zinc oxide in the bioplastic films was varied from 0.2%, 0.4%, 0.6%, 0.8% and 1.0% (w/w)	Harunyah <i>et al.</i> (2017)
	potato starch/nano-SiO ₂	80°C	30 mins	0.3% (w/v) nano-SiO ₂ of different sizes (15, 30, 80, and 100 nm) was added to 300 mL distilled water and then sonicated for 20 min to obtain a homogenous suspension. Subsequently, 5% (w/v of water) of potato starch and 5%(w/v of water) glycerin were added	Zhang <i>et al.</i> (2018)
Thermocompression	Thermoplastic corn starch with chitosan	140°C	5 mins	30% glycerol Starch chitosan microparticle content was varied from 0 to 50%	Paiva <i>et al.</i> (2018)
	2g of PLA was thermally compacted with a Carver laboratory press	190°C	3 mins		Rhim <i>et al.</i> (2005)

data. Fourier Transform Infrared (FT-IR) spectroscopy is commonly used to check the chemical structure and bonding that formed in the packaging film. In the IR spectrum, a broad band indicates hydroxyl (-OH) groups due to vibrational stretching while the narrow band is exhibiting stretching of the C-H bond (Chisenga *et al.*, 2019).

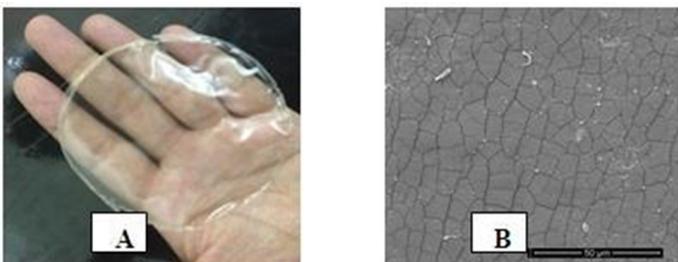


Figure 2. (a) Physical outlook of a pure biodegradable starch-based film. (b) SEM image of the starch-based film (Source: Wahyuningtiyas *et al.*, 2017).

In addition to that, physical appearance is also a factor to be concerned about in food packaging films. Most of the original films are transparent, however, the transparency of the packaging might be changed according to the types of food, light resistance, labelling on the packaging and design for attractive purposes.

3.2 Water absorption

Water absorption is an important factor for biodegradable composite materials for their application typically in the packaging industry. The hydrophilicity measurement of the polymeric film was evaluated by measuring the water absorption capacity of the film surface. Food packaging protects the food from being oxidized, reacting to water vapour, ultraviolet light, and both chemical and microbiological contamination. Therefore, food packaging contributes to the delay of the shelf-life and maintenance of the safety of the packaged food product. One of the methods to protect against food spoilage is to minimize water vapour or moisture in the packaging which may cause the growth of fungus or bacteria.

Figure 3 shows the water absorption capacity of arrowroot starch films and arrowroot starch films with different percentages of glycerol (Tarique, 2021). From the research team's result, as the amount of glycerol increases, the composite film shows better hydrophobic properties compared to the other two films. The major drawback of this starch/glycerol film is its poor tensile properties. An increase in glycerol content minimizes the water absorption capacity. On the other hand, the presence of more hydroxyl groups shows higher water absorption capacity as found in the starch/polyvinyl alcohol-based film. The starch/PVA film shows 227% water absorption capacity in 24 hrs. The increased amount of PVA will cause higher water absorption

capacity which is in the agreement with the review by Judawisastra *et al.* (2017) as in Figure 4. From their study, the amount of PVA up to 29wt% is directly proportional to the water absorption capacity while inversely proportional to the mechanical properties of the films, especially brittleness. Therefore, an optimum set of properties is required depending on the specific condition of the application.

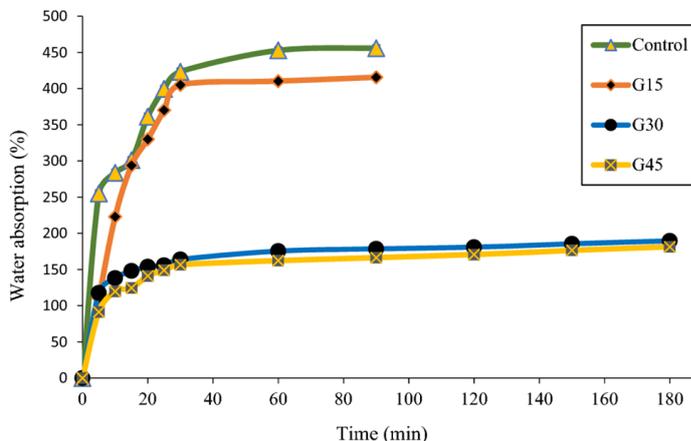


Figure 3. Water absorption of the control and G-plasticized AS films as a function of time (Source: Tarique *et al.*, 2021).

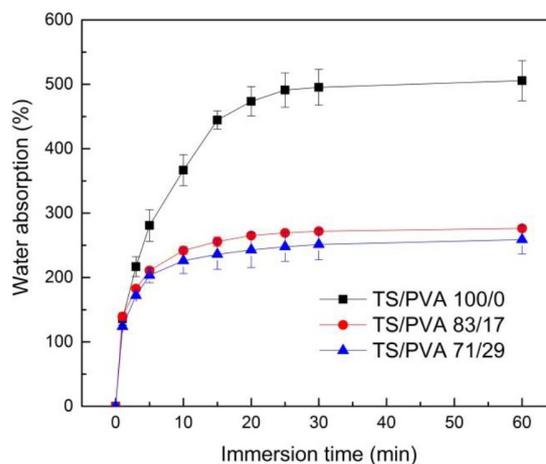


Figure 4. Water absorption of tapioca starch/ polyvinyl alcohol (TS/PVA) bioplastics (Source: Judawisastra *et al.*, 2017)

In the food industry, food is required to be kept dry or at a suitable level of moistness as certain food need specific moisture to maintain freshness such as green leaves vegetables, beans products and fresh mushrooms. The water absorption capacity of the packaging relates closely to the water vapour permeability, which is one of the factors of concern in biodegradable packaging films.

3.3 Mechanical properties

The tensile strength of the biofilms was significantly affected by the addition of thermoplastic chitosan processed by extrusion. The presence of thermoplastic chitosan reduced the tensile strength of the blends, which was probably because of their plasticizing capability. The addition of chitosan led to a reduction in elastic

modulus, thus forming fewer rigid films (Harunsyah and Fauzan, 2017). It has been reported that the addition of the plasticizer helps the thermoplastic chitosan matrix to become less dense, thus facilitating the movement of the polymer chains and improving the flexibility of the films. These results are consistent with the literature because of the increase in elastic modulus of the blends with respect to thermoplastic chitosan as displayed in Figure 5. This can be explained based on the presence of hydrogen bonds between the plasticizer and starch molecules (Mendes *et al.*, 2016).

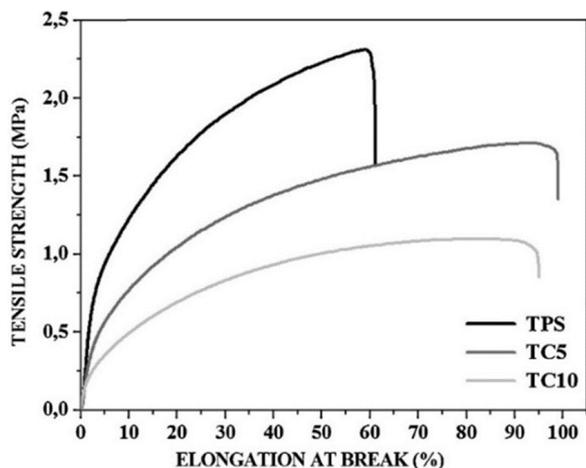


Figure 5. Stress-strain curve of plastic film with chitosan additive (Source: Mendes *et al.*, 2016).

The tensile properties of modified cellulose/PVA are presented in Figure 6. It is observed that tensile strength and Young's modulus of films increase as the percentage composition of the modified cellulose increases. However, a decrease in elongation at break is observed as the percentage composition of modified cellulose increases. With the increasing cellulose content, the interactions between the cellulose and the matrix are improved and crack propagation was inhibited, which resulted in the increased tensile strength and Young's modulus (Sandeep *et al.*, 2012). In contrast, the biodegradable packaging formed by chitosan/ PVA was proven high in tensile strength when chitosan was added

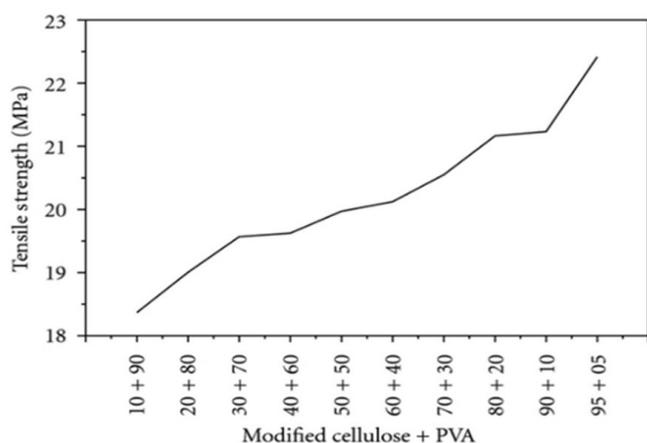


Figure 6. Tensile strength of modified cellulose/ PVA film (Source: Sandeep *et al.*, 2012).

to improve the crosslinking between OH groups in PVA (Hidayati *et al.*, 2021).

In terms of mechanical properties, the production of food packaging films concerns the tensile strength, elongation of break, brittleness and stiffness of the films. The packaging properties depend on the types of food to be packed. Generally, the food packaging films are required to be high in tensile strength and elongation of the break to support the food in the packages as the loads.

3.4 Antimicrobial properties

Incorporation of antimicrobial substances viz. cinnamaldehyde, lemongrass oil, peppermint oil, and clove oil was carried out in the polymer matrix material, to provide antimicrobial properties against the pathogenic and food spoilage bacteria namely *Bacillus cereus* and *Escherichia coli*. The mixture formulation of the biodegradable films incorporated with 0.5% level of cinnamaldehyde, lemongrass oil, clove oil, and peppermint oil were reported by Azahari *et al.* (2011). About 150 μ L of each solution was poured into a well of 10 mm on solid media in nutrient agar inoculated with *E. coli* and *B. cereus*. The antimicrobial effect of lemongrass oil incorporated into starch films was highest followed by clove oil. The peppermint oil incorporated films exhibited the lowest antimicrobial activity against both *E. coli* and *B. cereus*. Furthermore, it was observed that *E. coli* exhibited a lower zone of inhibition diameter than *B. cereus*, irrespective of the type of oil used to develop starch-based films. Lemongrass oil has the highest antimicrobial efficacy whereas peppermint oil has the lowest antimicrobial efficacy against test organisms. Besides oils or extracts from plants being good additives for antimicrobial purposes, nanoparticles also acted as antimicrobial agents. The benefits of nanoparticles in biodegradable packaging films will be discussed in the next section.

4. Nanoparticles as additives in biodegradable films

In the recent production of active food packaging, various kinds of metal oxide nanoparticles were added as additives. Nanoparticles such as zinc oxide (ZnO), titanium dioxide (TiO₂), and silicon oxide (SiO) have proven effective in antibacterial, enhanced mechanical properties, as pigment and more. Harunsyah and Fauzan (2017) synthesized bioplastic with zinc oxide nanoparticles by heating the mixture solution of starch and zinc oxide at 85 \pm 5 $^{\circ}$ C for gelatinization. It is proven the change of mechanical properties of the films with nanoparticles as an additive in the previous study not only for the bioplastic but also for other types of films such as regenerated cellulose film (Evyan *et al.*, 2017).

The tensile strength properties are affected by the concentration of glycerol as a plasticizer which increases the elongation and reduces the tensile strength of the bioplastic. Meanwhile, there was a study to produce biodegradable chitosan film with ZnO nanoparticles using food industry by-products (Souza *et al.*, 2021). The incorporation of ZnO generally increases the ductility and reduced the brittleness of the formed chitosan film by improving the elongation at the break while the lowered tensile strength of the formed films. The prepared film with ZnO as a plasticizer possesses properties that are suitable and high potential to be chosen in the application of food packaging which offers preservation of freshness.

In certain studies, the incorporation of the nano-SiO₂ significantly affected the tensile strength of the potato starch films. The matrix of the potato starch films could be strengthened by O–Si–O bonding, hydrogen bonding, or electrostatic attraction with the incorporation of nano-SiO₂ (Zhang *et al.*, 2018). The optimal tensile strength was obtained when the 100 nm nano-SiO₂ was added during the production of the film. The nano-SiO₂ was well adsorbed and bonded with potato starch in the formation of the films, thus improving the interphase adhesion strength between the matrix and the nano-SiO₂. In addition, the smaller nanoSiO₂ was more prone to agglomeration in the dispersion (Zhang *et al.*, 2018).

In 2012, there is a novel starch/PVA/TiO₂ biodegradable film preparation using the casting method (Hejri *et al.*, 2012). The addition of TiO₂ enhanced the thermal stability of the films due to the interfacial adhesion and compatibility of TiO₂ between starch/PVA matrices. The thermal stability of the packaging set a potential application that required high-temperature conditions. In another research project, Zhang's research team (Zhang *et al.*, 2018) synthesized biodegradable food packaging for strawberries with the addition of silver (Ag). The produced packaging work effectively with lowered vitamin C loss, lengthening the decline period of phenol and 1-Diphenyl-2-picrylhydrazyl (DPPH) in strawberries.

On the other hand, the films added with nano-SiO₂ of different sizes were tested for their antibacterial activity against *E. coli* and *Staphylococcus aureus* (Zhang, Wang and Cheng, 2018). The inhibitory effect was determined based on the clear zone around the film discs. All the nano-SiO₂/potato starch films were not infested with bacteria, indicating that external bacteria were prevented from passing through the films to infect food in the packaging process. The nano-SiO₂/potato starch films showed a significant inhibitory effect against *E. coli* and low efficiency against *S. aureus*. This could

be caused by adsorbing on the cell walls of the bacteria, destroying the structure of the cell membranes. The structure of the cell organelles was then destroyed because of the permeation of the nano-SiO₂. Thus, nano-SiO₂ could damage the antioxidant system and produce stronger lipid peroxidation (Walter *et al.*, 2018). The uniform dispersion of the nano-SiO₂ with small particles in the potato starch films plays a crucial role in fruit and vegetable preservation. Table 4 summarizes the additives of nanoparticles and plant oils or plant extracts in biodegradable food packaging films with the specific function based on the types of food that are to be preserved. Most of the additives benefit in terms of antimicrobial agent, antioxidant agent, and water vapour permeability.

5. Conclusion

Biodegradable films are the best choice for humans to reduce the usage of conventional plastics and minimize the pollution and side effects on humans and the environment. Although there are countless advantages to adding nanoparticles to the formation of biodegradable food packaging, there are still gaps and limitations when applying them. For an instant, the mixture of materials for bioplastic was less homogenous when the nanoparticle was added as observed by Wahyuningtiyas under SEM for the morphology of biodegradable plastics with the addition of ZnO by 1% - 6% (2017). The starch is suspected granules with other ingredients in the mixture. This might due to the addition of ingredients in the glycerol process that interacts with starch to form granules that are difficult to dissolve. Meanwhile, many are questioned about the safety of nanoparticles in food packaging. Seepage of nanoparticles may occur when using the food packaging containing nanoparticles and there is a possibility of allowing consumers take in a small amount of the nanoparticles.

It is difficult to claim that bioplastics are more environmentally friendly than traditional plastics when all aspects of their life cycle are considered land use, pesticides and herbicides, water use, greenhouse gas, energy consumption and methane emissions, and biodegradability, recyclability and more. However, as researchers around the world work to develop greener varieties and more efficient production processes, bioplastics do hold promise to help lessen pollution and reduce our carbon footprint.

Researchers have to consider the application and the choice of nanoparticles during the production of bioplastic. Government and relevant bodies should cooperate to enforce the use of biodegradable packaging and film since the usage of packaging cannot be avoided

Table 4. The additives in biodegradable food packaging based on types of preserved food.

Principal film	Benefits	Additives	Functionality	Preserved food	References
Polylactic acid (PLA)	Low cost Biocompatibility	AgNPs	- Antimicrobial agent	Strawberries	Zhang et al. (2018)
Cellulose	High in hydrophilic present free hydroxyl moieties, addition of - OH moieties to improve cross-linking Replace PE with biodegradable property	Clove and oregano oil	- Antimicrobial agent - Antioxidant agent - Sensory features	Iceberg lettuce	• Wieczynska et al. (2018)
Carboxymethyl cellulose with carrageenan		Grapefruit seeds extract	Water resistant	Rainbow trout fillet	• Hassanzadeh et al. (2018)
Chitosan	Most abundant natural polymer	TiO ₂	Antimicrobial agent	Tomato	• Kaewklin et al. (2018)
	Intrinsic antibacterial activity				•
	Antioxidant				•
	Biocompatible Mostly edible	Ginger oil	Antimicrobial agent	Poultry	• Souza et al. (2021)
PVA	Signal for improper storage condition in intelligent packaging	Satureja plant oil		Lamb meat	• Pabast et al. (2018)
	High in hydrophilicity	Limonene	Antimicrobial agent, oxygen scavengers	cucumber	• Maleki et al. (2018)
	Water solubility	Nanoparticles: Ag, ZnO, TiO ₂ , Fe ₃ O ₄	Improved WVP Antimicrobial agent Antimicrobial activity	Cheese	• Youssef et al. (2018)
	Good crystallinity with hydroxyl moieties at side chains (help in loading of additives via hydrogen & covalent bonding)	Essential oils	Antimicrobial agent Antimicrobial activity	Ground beef	• Talebi et al. (2018)
Gelatin	Water soluble protein	Thyme oil	Antimicrobial agent	Chicken	• Ejaz et al. (2018)
	Obtained from hydrolysis of collagen Edible	ZnO	Antimicrobial agent	Shrimps	• Ejaz et al. (2018)

in modern life. The pollution and issues that influence health and the ecosystem are becoming crucial thus the usage of biodegradable packaging, especially for food which is our daily consumption could be a vital step in moving toward sustainability.

Conflict of interest

The authors declare no conflict of interest.

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