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Article

Characterization of Polyvinyl Alcohol (PVA) as Antimicrobial Biocomposite Film: A Review

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Abstract— **Packaging is a critical process in the food industry because it is used to prevent spoilage, extend shelf-life, and provide an attractive presentation of the food product. Plastic packaging is used all over the world, and its production is increasing year after year. It comes in a variety of colours and designs. However, it has caused serious environmental problems, particularly to the ocean that has become a place for discarded plastic packaging. To address this issue, biodegradable packaging was developed to replace the use of plastic packaging because it helps to reduce environmental impact and waste management costs. Biodegradable packaging is also known as environmentally friendly packaging because it can be degraded into carbon dioxide, water, inorganic compounds, and biomass by microorganisms, algae, fungi, as well as enzyme catalysts. Biodegradable biocomposite film such as starch, cellulose, chitosan, and polyvinyl alcohol (PVA) is required to produce biodegradable packaging. Therefore, this paper aims to characterize PVA as a biocomposite film in biodegradable packaging. PVA has excellent properties to form films, as well as biodegradable, abundant in the environment, and cost-effective. However, it has some limitations in terms of thickness and mechanical properties; thus, the incorporation of PVA with essential oils and fiber is required to improve its mechanical properties, thickness, and provide antimicrobial properties to the packaging.**

Keywords— **Plastic packaging; Biodegradable packaging; Biocomposite film; Polyvinyl alcohol (PVA); Antimicrobial film**

I. INTRODUCTION

Packaging is now one of the most important items needed in daily life because it primarily serves to package items. Demand for packaging increased by about 5.2% year by year, reaching more than USD 300 billion in 2019 [1]. Packaging has become one of the most important items in the food industry because it is used to package food products to extend shelf-life and thus reduce food deterioration [2].

Petrochemical-based plastics derived from fossil resources are widely used because they have excellent heat resistance properties, as well as strength, stability, saleability, good barrier, permeability, flexibility, and rigidity [3]-[5][9]. Furthermore, plastic packaging is available in a variety of designs, lightweight, and inexpensive, but it has caused serious environmental problems [6] to nature due to its non-biodegradable properties. Once used, the ocean has become the most convenient place to dispose the plastic packaging [7]. According to data, approximately 8 million tonnes of plastic packaging escape into the ocean each year, primarily from developing countries due to the lack of space for garbage collection, an inefficient garbage system, and low recycling rates [8]. As a result, it has raised global concern among people because this packaging has limited disposal options, causing an increase in environmental issues such as greenhouse gas emissions and the safety of the marine environment [9]-[10]. Approximately, thousands of marine lives are threatened with extinction that indirectly reduced revenue for the fishing and tourism industries [11]. Non-biodegradable packaging has a complex molecule, making it difficult for microorganisms to break down [2]. As consumer concern grows, they demand packaging that can be destroyed and decomposed naturally in the environment [12].

Biodegradation refers to the ability of the material to undergo the defragmentation process during exposure to the natural environment with the help of microorganisms such as bacteria, algae, and fungi. The process cuts longer molecular compounds into smaller compounds, which causes the loss of mechanical properties or chemical modifications, resulting in the final products of carbon dioxide, water, inorganic carbon, and organic carbon [2][5][10][16]-[17]. The physiological system of plant will absorb gases that are released during the process [18].

Scientists discovered biodegradable packaging made of the natural or synthetic biodegradable polymer [13] to be the best alternative to plastic packaging. The biodegradable packaging consists of a biocomposite film that plays an important role in providing biodegradable properties, in which the film is moisture resistant and has an oxygen barrier to allow gases through the film [2][14]. Furthermore, polymer resources are easily obtained [14] because starch, cellulose, and chitosan are commonly used. Some properties of the biocomposite film such as renewable resources and polymeric matrices have made it the best option for packaging applications, which can reduce the greenhouse effect and preserve the environmental benefit [14]-[15].

PVA is a resin synthetic polymer with excellent properties for biocomposite film. It has a strong oxygen barrier, which allows small amounts of gas because the hydrogen bonding between the hydrogen groups is too strong [19]. PVA also has excellent properties in film formation and chemical resistance [20]. PVA is a polymer with a unique planar zig-zag structure with a carbon backbone [7][21]-[22]. On top of being biodegradable, PVA is also cost-effective, fully renewable, non-toxic, non-flammable, and non-ionic [23]-[24], which results in high tensile strength and elongation at film break [24]. PVA is synthesized from vinyl acetate via a radical polymerization process that yielded polyvinyl acetate (PVAc) as an intermediate product, which was then followed by a hydrolysis process on the acetate group of PVAc to allow the changes to the hydroxyl group to produce PVA [26]-[28]. There are two types of PVA hydrolysis based on the degree of hydrolysis; fully hydrolyzed and partially hydrolyzed PVA [13]. The temperatures involved in the fully and partially hydrolyzed processes are 230°C and 180 to 190°C, respectively [29]. The degree of hydrolysis is determined by the number of residual acetate groups that did not occur during saponification or alcoholysis [13]. Because of the pyrolysis process that occurs at high temperatures, it can decompose rapidly at 200°C [28]. However, PVA has some limitations that must be considered such as a low rate of biodegradability due to the carbon-carbon linkages on its backbone. Therefore, the incorporation of PVA with nanocomposites is required to increase the rate of biodegradability as well as to improve the film thickness and mechanical properties [15][17].

Based on the properties of PVA, it is a promising biocomposite film for biodegradable packaging. Therefore, this paper aims to characterize PVA as a biocomposite film in biodegradable packaging.

II. PREPARATION OF POLYVINYL ALCOHOL

There are many different types of PVA available, ranging from commercial, low molecular mass PVA to lab-scale PVA. Each type of PVA necessitates a unique monomer and medium. However, the process is commonly known as polymerization. The preparation of PVA is shown in Table I that includes the monomer, process, and medium used.

Table I the preparation of PVA according to the types of PVA

Types of PVA	Monomer	Process	Medium	Reference
Commercial PVA	Vinyl acetate	Saponification	Aqueous sodium hydroxide	[27]
		Hydrolysis	Methyl alcohol of strong base powder	
	PVAc	Hydrolysis/ Polymerization	Methanol and sodium hydroxide	
Low molecular mass PVA	Acetaldehyde	Polymerization	Low temperature (-80°C to -20°C) with amalgam	
Lab scale PVA	Vinyl tert-butyl ether	Polymerization	Boron trifluoride diethyl etherate in toluene at the temperature of -78°C	
			Methylene chloride	
	Poly (vinyl trimethylsilyl ether)	Polymerization	Ferric chloride in nitroethane of -78°C	

As shown in Table I, there are three types of PVA preparation available. To begin the process, each type of PVA used a different monomer. There are also various mediums used during the process depending on the monomer and the process to be carried out. Since vinyl monomer is unstable, PVAc is used in the polymerization process [30]. This process will produce repeated structural units that are not monomer-based [13]. Hermann and Haehnal (2014) discovered a continuous process using the medium of ethanol and potassium hydroxide to enable the hydrolyzation of acetate group PVAc by ester interchange at 80°C for 40 min [29].

Commercial PVA can be made either using vinyl acetate or PVAc as the monomer in a variety of processes and mediums. There are two types of PVA based on the degree of vinyl acetate hydrolysis the duration of the process, which are fully hydrolyzed PVA and partially hydrolyzed PVA [13]. Commercial PVA has molecular weights ranging from 30,000 to 200,000 g/mol [13]. The higher the molecular weight, the larger the crystalline structure [31], which contributes to various properties such as pH, melting point, and refractive index. Furthermore, the physical and functional properties of fully and partially hydrolyzed PVA are different. Fully hydrolyzed PVA is soluble in water but insoluble in an organic solvent, while partial hydrolyzed PVA is soluble in the organic solvent but insoluble in water [27].

To date, low molecular mass PVA is available in the market, in which the manufacturing process requires acetaldehyde to be polymerized at low temperatures in the presence of amalgam.

There was also a lab-scale PVA that used vinyl tert-butyl ether and poly (vinyl trimethylsilyl ether) in the polymerization but with different mediums depending on the monomer as shown in Table I.

III. INCORPORATION OF PVA WITH ESSENTIAL OILS

Table II shows the incorporation of PVA with essential oils such as *Laurus nobilis*, *Rosmarinus officinalis*, *Origanum vulgare*, and *Zataria multiflora* to improve some of the properties of PVA film.

Table II the incorporation of PVA with essential oil

Types of Essential Oils	Concentration of Essential Oils	Concentration of PVA	Reinforcing agent	Method	Reference
Laurus nobilis	10% w/w	14% w/v	NA	Electrospinning	[33]
Rosmarinus officinalis	10% w/w	14% w/v	NA	Electrospinning	[33]
Origanum vulgarae	2% v/v	2% w/w	Alpachitin nanocrystals 5% w/v	Solvent casting	[34]
Zataria multiflora	10% w/v	2% w/w	NA	Emulsification	[22]

As shown in Table II, several types of essential oils from the spice category have been discovered to be incorporated with PVA. PVA has hydrophilic properties that make it easy to degrade via the hydrolysis process due to the presence of hydroxyl group at the repeating unit [37]. Therefore, essential oils are commonly used to incorporate with PVA due to their lipid structure that can reduce the water vapor permeability in hydrophilic materials [36]. This addition may also improve the structural, mechanical, and optical properties of the film [36]. The incorporation of essential oils with nanocomposites may aid in preventing the loss of volatile essential oils during the handling, heating, and oxygen exposure processes [33].

The electrospinning process is used to combine PVA with essential oils of *Laurus nobilis* and *Rosmarinus officinalis*. It produces a complex hierarchical structure that requires specific conditions such as a longer production cycle, a specific temperature, and a specific pressure condition [38]. Citric acid is occasionally used as a crosslinking agent during the incorporation process. It is a natural crosslinking agent that can keep PVA intact longer than other crosslinking agents [32]. It encourages esterification and crosslinking, which improves the mechanical, structural, and barrier properties of the film [39]. It also acts to balance the hydrophilicity of polymer networks, as well as to provide hydrogen bonding and additional binding sites [37].

There are two types of cross-linking that are commonly used in food packaging, which are physical or chemical processes. However, their side effects have become a concern because they may cause cytotoxicity due to the leaching of the agent from the packaging into food products. [32]. In addition, the cross-linking agent can reduce crystallinity, as well as improve mechanical properties and the interaction of hydroxyl groups with oxygen-containing groups [37].

The incorporation of PVA with fibre improves mechanical and thermal properties [18]. Banana pseudostem, and pomegranate fibers were studied for their ability to incorporate with PVA using the casting method. Citric acid was used as a crosslinking agent in PVA/Banana pseudostem to improve the water-resistance properties of the film [40]. The biocomposite film has higher compatibility due to the formation of hydrogen bonds as a result of this incorporation [40].

The electrospinning method was then used to incorporate PVA/D-Limonene. D-Limonene was extracted from orange peel oil. During the electrospinning process, the PVA powder caused the PVA to be ionized by deionized water at room temperature, which allowed the swelling process to occur and the addition of D-Limonene solution with ultrasound treatment in an ultrasound liquid processor. As a result, fiber properties and uniformity were improved [41].

In film preparation, 1.94% of PVA was combined with 2 cm pomegranate peels and essential oils such as clove and thyme; however, no cross-linking or reinforcing agents were used [41].

IV. PROPERTIES OF PVA FILM INCORPORATED WITH ESSENTIAL OILS

The incorporation of PVA with essential oils was evaluated based on the mechanical properties of the film. Table III shows the effect of incorporation on tensile strength, elongation at break, and thickness of PVA film.

Table III properties of the biocomposite film after incorporation

Type of essential oils	Tensile strength (MPa)	Elongation at break (%)	Thickness (μm)	Reference
Laurus nobilis	NA	NA	NA	[32]
Rosmarinus officinalis	NA	NA	NA	[32]
Origanum vulgarae	13.06±3.85	298.97±85.50	7.28±2.0	[33]
Zataria multiflora	13.5±0.61	216±4	NA	[22]

Tensile strength refers to the tensile stress of the film. It is sustained at the maximum rate to maintain its integrity during the tension test and the deformation value is adjusted according to the demands [22], which implied stretching capacity [43]. The better the biocomposite film, the higher the tensile strength. Meanwhile, elongation at break refers to the film's ability to be flexible and extensible to breakage [22], which refers to the measurement of the film stiffness [43]. Tensile strength and elongation at break are mechanical properties that can be measured by Instron Universal Testing using standard ASTM D 882-88 methods [86][34]. The thickness of the film can be determined using a digital micro meter [34].

The addition of essential oil to PVA films results in a decrease in tensile strength and an increase in elongation at break [22]. This is because the strongest polymer-polymer interaction is weakened as it is replaced by weak polymer-oil interaction. The elongation at break increases as the film flexibility increases while the thickness of the film decreases as the film lightness decreases [34].

The mechanical properties and thickness of the PVA/Pomegranate fiber film were not available for fiber incorporation. For PVA/D-Limonene, the tensile strength and elongation at break was 3.87±0.25 MPa and 55.62±2.93%, respectively [41]. In general, incorporating PVA with fiber increases tensile strength while decreases the elongation at break. However, the tensile strength and elongation at break for PVA/Banana pseudostem decreased as the banana content increased, with elongation at break were greater than 100% and tensile strength of 30.8 MPa when using 20% of banana fiber.

V. ANTIMICROBIAL PACKAGING

Antimicrobial packaging contains antimicrobial agents such as nanoparticles, ethanol, essential oils, and organic acids [44], which helps to extend the shelf-life of food products by inhibiting the growth of pathogenic bacteria and preventing food spoilage [44]-[45]. There are two types of antimicrobial packaging available. The first type is packaging with direct contact between the antimicrobial surface and the food products. The second type is packaging with indirect contact between the antimicrobial surface and the food product [1].

Since PVA film lacks antimicrobial, antioxidant, and antifungal properties, the incorporation of PVA with essential oils demonstrates that the packaging can exhibit excellent functional properties [22]. A previous study showed that the incorporation of PVA with clove oil could inhibit bacteria growth and lipid oxidation of the *Trichiurus haumela*, in addition to antioxidant activity in the vapor phase [34]. The essential oils obtained from the extraction of aromatic compounds contain antimicrobial and antioxidant activity [46], which is beneficial to suppress the pathogen growth and the occurrence of lipid oxidation [34][46]. Acids, alcohol, aldehyde, and terpenes are among the active components found in essential oils that aid in antimicrobial activity [47]. Crosslinking agents are also used to aid in the performance of the bactericidal activity and the physicochemical properties of the film [8]. As a result, the quality and safety of the packaging are preserved [48].

The procedure to produce antimicrobial packaging starts by adding an antimicrobial agent to the film-forming solution and observing its behavior during the casting phase [49]. The behavior is determined by the nature of the antimicrobial molecules. The antimicrobial agent in the film will then be moved and diffused to the phase in contact with a low temperature [49].

The benefits of antimicrobial packaging include the prevention of foodborne pathogen outbreaks, as well as the reduction of food product losses and waste due to the ability of the antimicrobial agent to suppress the growth of microorganisms [50]-[51]. As a result, the food product is of high quality and safe [49].

The use of antimicrobial packaging is proven to be effective in slowing the growth of spoilage and pathogenic microorganisms in contaminated foods due to the modification of the active environment, as there is a constant interaction between the food and the specified shelf life [52]-[53]. The antimicrobial agent also inhibits the membrane structure and essential microorganism pathways [54].

VI. BIODEGRADABILITY OF PVA FILM

The biodegradable packaging is degraded in either aerobic or anaerobic conditions [55]. The aerobic process includes soil composting, biological drying, and some aquatic environments, while the anaerobic process includes anaerobic digestion, methane fermentation, and a few aquatic habitats [55]-[56]. Biodegradation of the biocomposite film by soil microorganisms results in the production of water, carbon dioxide, and monomer [34]. The biodegradability of the materials is determined by their chemical composition, bonding nature, and water availability [33]. Polymer properties and composition, as well as climate, humidity, and atmospheric pollutants have significant impacts on biodegradability [16]. Polymer biodegradability is also influenced by its structure, morphology, chemical treatment, and molecular weight [13].

The biodegradation process is divided into three stages, which are bio-deterioration, bio-fragmentation, and assimilation. Bio-deterioration involves the polymer change on the chemical, mechanical, and physical properties due to the biological activity of the microorganisms on the surface material [56]. Bio-fragmentation involves the conversion of polymers into oligomers and monomers from the breakdown process of microbial activity. The final stage is assimilation, in which microorganisms cause bio-fragmentation to the biodegradation of carbon dioxide, water, and biomass [56].

PVA has biodegradable properties with an average molecular weight of 106 g/mo. It can be degraded by 55 species of microorganisms, including bacteria, fungi, and algae, with *Pseudomonas* being the main degrader in PVA degradation [13][17]. PVA has a carbon backbone due to the occurrence of the enzyme catalyzed-oxidation process at the functional group during the process of microbial utilization for the carbon source [13]. The presence of hydroxyl groups on the carbon atoms is responsible for the degradation of PVA via hydrolysis mechanism [13].

Thus, PVA is lightweight, cost-effective, and has a wide range of physical and chemical properties [58]-[60]. The biodegradable properties of PVA can be an effective solution for the environmental problems, particularly the greenhouse effect, soil fertility, waste management costs, the number of discarded plastics in the environment, and the fatal risk to wild and marine animals [57][59].

VII. CONCLUSION

The current review reports the potential of PVA in biodegradable packaging as the best alternative to plastic packaging. Plastic packaging is non-biodegradable that contributes to environmental pollution. Therefore, the use of biocomposite film in packaging applications becomes the highlight to produce biodegradable packaging. PVA is a synthetic resin polymer derived from the radical polymerization of vinyl acetate. PVA is used as a biocomposite film because it is abundant in the environment that can reduce waste management costs. On top of that, it has excellent biodegradability and film-forming properties. Furthermore, the degradation process of PVA is carried out naturally mainly by *Pseudomonas*, which is catalysed by an enzyme. Thus, the disposal of this PVA packaging could increase soil fertility. The review also highlighted some limitations in the applications of PVA packaging. To address the issue, PVA should be combined with either essential oil or fiber to improve its mechanical properties and increase its biodegradability.

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