A Review on PVA Based Biodegradable Films: A New Hope for Plastic Pollution Remediation

Nanda Karmaker,[1-3] Hironmoy Karmaker,[2] Ruhul A. Khan*[3]

[1]Department of Applied Chemistry and Chemical Engineering, University of Dhaka, Dhaka-1000, Bangladesh.
[2]Department of Mechanical Engineering, Rajshahi University of Engineering and Technology, Rajshahi, Bangladesh..
[3]Polymer Composite Laboratory, Institute of Radiation and Polymer Technology, Atomic Energy Research Establishment, Savar, Dhaka-1349
Corresponding author: dr.ruhul_khan@yahoo.com

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ABSTRACT

With the progress of modern science, people have started paying attention to degradable film. Biodegradable film not only has the function and characteristics of traditional synthetic polymers, but also it will revert to environment. These film attains the purpose of protecting the environment by using degradable materials such as agar, carrageenan, chitosan, gelatin, sodium alginate, etc. Polyvinyl Alcohol (PVA) is a water-soluble semi-synthetic polymer. This hygroscopic nature of PVA adds a new dimension to the world of biodegradable films. In this modern civilization with the growing population, the need of packaging material is huge in every sector. The renowned 'Solution Casting" method is widely used for making these types of biodegradable films. In this process the core ingredients are mixed by their wt/v% ratio to make a desired composition of the films. After the fabrication the films are characterized by several mechanical and surface morphological techniques to understand whether the films are feasible, durable and sustainable mostly its impact on nature.

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Long term use of synthetic packaging is harmful for health as well as has an adverse effect on nature due to difficulty in disposal of garbage. So this paper conducts a clear exposition how PVA has open a wide wing to preserve our ecology accompanied by the other natural biopolymers.

INTRODUCTION

At the present time, the growing environmental pollution is caused by different factors, among which one of the most noteworthy is the accumulation of the synthetic polymer wastes produced by packaging. Thus, many researchers are involved in a creation of new biodegradable materials, which can be decomposed under environmental conditions with the formation of harmless-for-natural compounds. Such systems are of excessive awareness not only because of the lessening of environmental problems, but also because of the consumption of a non-renewable raw material such as petroleum, which is progressively rare and difficult to extract and refine (Thomas, 2012 & Leja, 2010).

The natural polymers such as collagen, starch, chitosan, keratin, gelatin, agar, carrageenan, alginate and silk can be converted into soluble derivatives by chemical and enzymatic hydrolysis. The hydrolyzed proteins, after characterization, can be used to design a polymeric matrix. A new method of preparation of polymeric materials for biomedical applications is the mixing of two or more polymers. Throughout the last three decades an increasing interest in new materials based on the blends of two or more polymers can be witnessed. Several laboratories have also been working on the progress of new materials based on the blends of two or more polymers and inorganic nano particles. New materials based on the blends of two polymers or biopolymers which contain nano particles can be used as a transplant in the context of both hard and soft tissues. Moreover, several documents have been published so far in the area of biomaterials based on blends of natural polymers and synthetic polymers comprising inorganic nanoparticles (Kasirajan, 2012; Kumar, 2009; & Pandey, 2005).

Development of eco-friendly packaging materials is still a stimulating area and many studies have been focused on the improvement of PVA mechanical and barrier properties by combination with other polymers or fillers in order to use it in the packaging industry. For many applications, mechanical properties of PVA should be considerably improved without damaging its valuable properties. Many studies highlighted the effectiveness of natural fibers in improving mechanical properties of PVA. Consideration has been focused on polyvinyl alcohol (PVA) for more than 40 years due to its unique chemical and physical properties as well as its

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industrial implements (Wu, 2006; Hablot, 2008 & Gutiérrez, 2017). This review defines some of the most studied biopolymers which may have a great importance in the future packaging industries. The aim is to cover current developments with respect to their prospective for use in the sustainable and green packaging industry. Especially the barrier properties are recognized as crucial issues for actual operation of bio based materials.

TYPES OF POLYMERS

The word “Polymer” is derived from two Greek words, ‘Poly’ that means many (numerous) and ‘Mer’ which means units. Simply stated, a polymer is a long-chain molecule that is composed of a large number of repeating units of identical structure. On the basis of its source of origin, polymers can be classified into three categories. Natural Polymers Natural polymers are polymers which occur in nature and are existing in natural sources like plants and animals. For example, cellulose, protein, rubber, seaweeds, etc.
Synthetic Polymers Synthetic polymers are polymers which humans can artificially create/synthesize in a lab. Polyethylene, Nylon etc are some of the synthetic polymers. Semi-Synthetic Polymers Semi-Synthetic polymers are polymers obtained by making modification in natural polymers artificially in a lab. Example: cellulose acetate (Rayon), poly vinyl alcohol (PVA), Poly Lactic Acid (PLA), etc. (Pokrovskii, 2009 & Roiter, 2005).

NATURE OF PVA FILMS

Manmade poly(vinyl alcohol) (PVA) films and fibers are polymer materials which are identified to biodegrade when disposed in atmosphere. This is recognized to the hydrophilic property of PVA. In spite of a number of published papers on PVA, the thermal and mechanical properties show much smaller change in crystallinity with respect to temperature in comparison with poly olefin polymers such as polyethylene and polypropylene. Moreover, it can withstand high temperature which is also notable for its selection. It is used in papermaking, textiles, and a variety of coatings (Lee, 1999; Alexy, 2003 & Yoon, 2007).

Preparation of PVA: PVA is not obtained by polymerization of the resultant monomer. The monomer, vinyl alcohol, is unstable regarding acetaldehyde. At first, polymerization of vinyl acetate is done and the resulting poly vinyl acetate is transformed to the PVA. Sometimes, precursor polymers are used, with formate, chloroacetate groups instead of acetate. The conversion of the polyesters is usually conducted by base-catalysed transesterification with ethanol is goven below (Shin, 2006).

$[CH2CH(OAc)]n + C2H5OH \rightarrow [CH2CH(OH)]n + C2H5OAc$

Structure of PVA: PVA is an atactic polymer that has crystalline properties. It is mainly 1,3-diol linkages [-CH2-CH(OH)-CH2-CH(OH)-]. Sometimes a few percent of 1,2-diols [-CH2-CH(OH)-CH(OH)-CH2-] is found, relying on the conditions for the polymerization of the vinyl ester precursor (Cheng, 2007).

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Physical Properties: Poly(vinyl alcohol) (PVOH, PVA) is a water-soluble semi-synthetic polymer. It has the idealized formula $[CH2CH(OH)]n$. It is white (colourless) and odorless. It is sometimes supplied as beads or as solutions in water. Density of PVA is 1.19-1.31 g/cm3 (Shi, 2008). Polyvinyl alcohol has outstanding film forming, emulsifying and adhesive properties. It is also unaffected by oil, grease and solvents. It shows high tensile strength and flexibility, as well as high oxygen and aroma barrier properties. Though these properties are reliant on humidity. So, with higher humidity more water is absorbed. Water acts as a plasticizer, helps to decrease its tensile strength, but increase its elongation and tear strength (Vanin, 2005 & Tharanathan, 2003) .

Thermal Properties: PVA has a melting point of 230 °C. It can be fully hydrolyzed within temperature range of 180–190 °C. It decomposes rapidly above 200 °C as it can undergo pyrolysis at high temperatures (Mahdavinia, 2016).

ROLE OF PVA AS BIODEGRADABLE FILM

Polyvinyl alcohol (PVA) is a vinyl polymer comprised with only carbon–carbon linkages. The linkage is the similar to those of typical plastics such as polyethylene, polypropylene, and polystyrene, and of water-soluble polymers such as polyacrylamide and polyacrylic acid. Among the vinyl polymers manufactured industrially, PVA is the only one recognized to be mineralized by microorganisms. It is non-toxic and non-hazardous. PVA is water soluble and biodegradable; hence it is highly recommended for making water-soluble and biodegradable carriers, which may be beneficial to the production of delivery systems for chemicals such as fertilizers, pesticides, and herbicides (Watts, 2001 & Yang, 2016).

ANALYSIS REGARDING COMBINATION OF PVA WITH OTHER BIOPOLYMERS

PVA-Agar: Agar is obtained from red sea-algae, which is known as agarophytes, and belong to the Rhodophyta. Mainly it is a mixture of two constituents, the linear polysaccharide agarose, and a heterogeneous mixture of smaller molecules called agaropectin (Thomas, 2015). Agar shows hysteresis, melting at 85°C and solidifying from 32–40°C. This is a proper balance between easy melting and good gel stability at relatively high temperatures. This property is much suitable for film formation. Agar is very useful for making edible films for the vegans (Duckworth, 1971). Agar-PVA film reveals outstanding thermo-mechanical properties. The melting temperature of film is higher than agar and PVA. The TS, Eb values are pretty enough to use this film as a packaging material. Surface morphological tests are very significant for this type of films (Wu, 2009 & Lyons, 2009). PVA-Carrageenan: Carrageenan is mainly originated from the family of linear sulfated polysaccharides that are extracted from red edible seaweeds. Carrageenans can be classified into three categories on the basis of degree of sulfation. There is one sulfate group per disaccharide present in Kappa, whereas iota-carrageenan has two and lambda-

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carrageenan has three. Carrageenan is soluble in hot water instead of cold water. It can be used as thickening or gelling agent in food and cosmetics industries. It is also a great alternative to gelatin due to its vegan source (Moshe, 2016 & Nickerson, 2010).

Carrageenan along with PVA shows excellent thermo-mechanical properties. Mechanical tests such as Young's Modulus, Eb and TS etc can be used to measure its mechanical property. The outcomes of these results are the proof of its stable mechanical properties and how they can be used to make biodegradable packaging materials. The sulphate group in carrageenan acts as a crosslinking agent to the PVA, which is ensured by surface morphological tests. Both of the polymers have high melting temperature, so the film shows heat resistivity to a great extent (Chen, 2002 & Meng, 2018).

PVA-Chitosan: Chitosan, a linear polysaccharide composed of casually distributed β-(1→4)-linked D-glucosamine (deacetylated unit) and N-acetyl-D-glucosamine (acetylated unit). It is produced by treating the chitin shells of shrimp and other crustaceans with an alkaline substance, like sodium hydroxide. It has melting temperature above 80oC. Chitosan has anti-bacterial properties and medicinal uses (Tonny, 2014; Bourtoom, 2009 & Alam, 2008).

Chitosan has great antimicrobial properties, which is very beneficial for medicinal uses. It also shows great echanical properties along with PVA. The TS, TM, Eb values are good enough to use this combination as packaging materials. Moreover the acyl groups present in chitosan and the C-C bonds in PVA also give outstanding bonding between the molecules which can be easily visualized by inspecting the surface morphological tests. Due to presence of PVA in films, they exhibit some crystalline property which is ensured by the XRD test. Chitosan and PVA film also shows excellent thermal properties. Treating the Chitosan film with sodium hydroxide (NaOH), can make the films insoluble (Abraham, 2016; Dey, 2014 & Haque, 2005).

PVA-Gelatin: Gelatin is mainly an irreversibly hydrolyzed form of collagen, whereas the hydrolysis reduces protein fibrils into lesser peptides; liable on the physical and chemical methods of denaturation, the molecular weight of the peptides decreases drastically within a broad range. It is luminous, colorless, flavorless food ingredient, resulting from collagen taken from subconscious body parts. Melting temperature of gelatin has a range over 120oC (Dey, 2014 & Chen, 2017).

Gelatin is mainly animal sourced biopolymer and it is edible. So, gelatin contributes into making edible film which is also a great source of protein to human being. Gelatin is a large molecule and combination of various types of proteins. It goes well with PVA to make biodegradable films which is of course ecofriendly. Gelatin-PVA film exhibits excellent mechanical and thermal properties (Ma, 2017 & Tao, 2018).

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PVA-Sodium Alginate: Sodium alginate is the sodium salt of alginic acid. Its empirical formula is NaC6H7O6. Sodium alginate is a gum, extracted from the cell walls of brown algae. It is white to yellow in colour and fibrous powder. It also shows hygroscopic nature and melts at above 300oC. Sodium alginate has antibacterial and medicinal properties. Sodium alginate has gelling, thickening, stabilizing and film forming properties. For this, it is widely used in food industry, textile industry, paper industry, dental Industry, cosmetic Industry and in water treatment processes (Huang, 2015 & El-Ghaffar, 2012).

PVA and Sodium Alginate have great combination which possess excellent thermal, chemical, morphological and mechanical properties. For some physical testing water content, swelling properties are determined which indicates a diversified range and much suitable for polymeric packaging. Surface morphological testing is done by Scanning Electron Microscopy (SEM). Sodium alginate exhibits some crystalline properties which can be evaluated by X-Ray Diffraction (XRD). Sodium Alginate and PVA both have high melting point. For this reason, the combined film shows good thermal properties. Differential Scanning Calorimetric process is much helpful to for evaluating thermal properties. Lastly, PVA-Sodium Alginate film shows good mechanical properties such TS, TM and Eb (Qi, 2015; Park, 2017; Martínez-Gómez, 2017 & Yang, 2014).

PVA-Cellulose: The most abundant organic polymer on Earth is cellulose. The presence of cellulose in cotton fiber is 90%, that of wood is 40–50%, and that of dried hemp is approximately 57%. It is a polysaccharide containing a linear chain of several hundred of β(1→4) linked D-glucose units. It appears to be white in color and found in coarse powdered form. Melting temperature of cellulose varies from 260-320oC, depending on the source of cellulose (Cai, 2009 & Yousefi, 2011).

Cellulose is tasteless, odorless, hydrophilic, insoluble in water and most organic solvents and is biodegradable in nature. Cellulose mainly contributes to paper industries. It is also converted into a wide variety of derivative products such as cellophane and rayon which are very useful for other chemical treatments (Peresin, 2010 & Carlsson, 2012). Cellulose goes well with PVA for biodegradable film preparation. The high molecular weight and melting temperature contribute to great thermo-mechanical properties. Some researchers have done many experiments and found outstanding results. Cellulose also shows great chemical changes along with PVA which is proven by Nuclear Magnetic Resonance Spectroscopy (NMR) and Scanning Electron Microscopy (SEM). As cellulose is insoluble in water, the water resistance properties are satisfactory (Zheng, 2014 & Abdulkhani, 2013).

PVA-Starch: Starch or amylum is a polymeric carbohydrate consisting of a large number of glucose units joined by glycosidic bonds. It is the most common carbohydrate in human diets and is contained in large amounts in

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staple foods like potatoes, wheat, maize (corn), rice, and cassava. Pure starch is a white, tasteless and odorless powder that is insoluble in cold water or alcohol. It consists of two types of molecules: the linear and helical amylose and the branched amylopectin. Depending on the plant, starch generally contains 20 to 25% amylose and 75 to 80% amylopectin by weight (Sin, 2011 & Follain, 2005). Starch is insoluble in water and gelatinization forms with cold water at 55oC. Mixing most starches in warm water produces a paste, such as wheat paste, which can be used as a thickening, stiffening or gluing agent. This nature works as a binding agent and contributes efficiently into film making procedures (Salleh, 2015).

Many research works were done to measure the compatibility between PVA and starch. They found the thermo-mechanical properties very useful to use this combination for biodegradable film. Water uptake properties decrease with the increase in percentage of starch (Parvin, 2010 & Salleh, 2017).

PVA-Glycerol: Plasticizers are additives which increase the plasticity and decrease the viscosity of a material. These are added to alter the physical properties. They drop the attraction between polymer chains to make them more flexible (Pal, 2006). Glycerol also unknown as glycerin, is mainly a polyol compound. It is colorless, odorless and viscous liquid. It has sweet taste and no toxicity (Dicharry, 2006).

PVA shows outstanding properties along with glycerol. Glycerol combines very well and worthy cross-linking which is experimented by many researchers. The presence of glycerol significantly affect plasticizing effect on PVA by reducing both the glass transition and melting temperature of PVA. Moreover, glycerol helps to reduce the hardness as well as elastic modulus (Mohsin, 2011).

PVA-Urea: Urea, also recognized as carbamide, an organic compound with chemical formula CO(NH2)2. It has two –NH2 groups linked by a carbonyl (C=O) functional group. It is whitish in color and found in coarsely granule form. It is readily soluble in water and melts in the range of 133-135oC. It is non-toxic and most abundantly used as fertilizer (Liu, 2017). Urea is a complex plasticizer. It is used along with ethanolamine so that the urea doesn't separate out from PVA. Urea can effectively modify the thermal and chemical properties of PVA film. It goes amazingly as a strong hydrogen bond is formed between hydroxyl group of PVA and amide group present in urea (Jiang, 2012).

CONCLUSION

Nowadays great concerns are being given to make existing non-degradable polymers biodegradable either by chemical modification or by the inclusion of additives (e.g. sensitizers and biopolymers). Degradable films play a vital role in this area of food preservation, because of its environmental safety and preservation. Initially, a new material can be

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expensive, but in large-scale production these biodegradable polymers can be used for applications such as packaging, diapers, disposable tableware and mulch films, biomedical purposes, etc. PVA is semi-synthetic and can go very well while combining with biopolymers or plasticizers. PVA goes simultaneously along with biopolymers and plasticizers. Although PVA has water-soluble nature but due to its excellent properties and availability, it can be an inexpensive source of raw materials. However, many woks have been done already but more research is needed to improve its properties.

FUTURE SCOPE

The next step of this study is to evaluate a comparative study and choose some best biopolymers which combined well with the PVA for an outstanding biodegradable films. Moreover, properties and scopes of various synthetic but degradable biopolymers such as poly (caprolactone) (PCL), poly lactic acid (PLA), etc. can be studied.

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