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Biodegradable Future: A Comprehensive Survey on the Development and Characterization of Starch-Based Films for Sustainable Packaging

Suhail Ahmad Bhat[1], Ashok Kumar[1], Tanuja Srivastava[1], Riti Jindal[1]
[1]Department of Food Technology, Bhai Gurdas Institute of Engineering and Technology, Sangrur

Abstract: This review is focused on the analysis of film-forming ability of starch employed from various sources like cereals, tubers etc. and the relationship between their structure and physio-chemical properties. According to different studies, casting method for starch based biodegradable film forming is preferable due to its greater feasibility to simulate the industrial process. To increase the workability and flexibility of starch based biodegradable films, addition of different components required such as lipid, plasticizers, polyols, hydro-colloids, fillers, or active compounds.

Keywords: Biodegradable films, Starch, Plasticizers, Fillers.

Research Paper

***Corresponding Author:**
Suhail Ahmad Bhat
Department of Food Technology, Bhai Gurdas Institute of Engineering and Technology, Sangrur

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INTRODUCTION

Biopolymers are the polymers which originate from natural resources like starch, protein, polysaccharide, lipids and resins. Starch is a widely used agricultural raw material for biodegradable film manufacturing, since it is inexpensive, easy to handle, totally biodegradable and abundantly available in nature [1]. It can be extracted from different sources like cereals, roots, tubers, palms and pseudo cereals such as amaranth and quinoa [2]. Starch is a renewable bio-polymer consisting of amylose and amylopectin [3].Amylose is a linear polymer of α-1, 4 anhydroglucose units that has excellent film forming ability, resulting strong, isotropic, odorless, tasteless, and colorless film [4]. Parallel to which, amylopectin is a highly branched polymer of short α-1, 4 chains linked by α-1, 6 glyosidic branching points occurring every 25–30 glucose units [5]. Hence, the branching points of amylopectin and the amylose leads to the formation of amorphous regions, while the short-branched chains in the amylopectin act as the main crystalline components in granular starch, the higher content of amylopectin in native starch means greater crystallinity [6]. Amylose/amylopectin ratio depends on the source and age of the starch. Starch generally contains 20 to 25% amylose and 75 to 80% amylopectin [7]. Depending upon the origin, starch molecules may vary in their shape, size, structure, and chemical composition e.g. the *Chenopodium* seed (*Chenopodiumalbum*) is a small grain (1-3 mm diameter) which has an amylose content of ∼10–21% (depending on the variety) and a small starch granule size (∼1 μm). Small size of starch granule is one of the characteristics that allow for easier dispersion which makes the starch a promising material for film production [8].

Different types of film formation can take place and it can be composite or non-composite in nature. The features of the resulting film will depend on (1) the natural interactions taking place between starch, proteins, and lipids during drying of the filmogenic solution; (2) the distribution of the interactions within the polymeric matrix; (3) the balance between hydrophilic and hydrophobic interactions; and (4) the concentration of each component in the film. In addition, starch is able to form transparent edible biodegradable films without any previous chemical treatment **[**9**]**. On the other hand, in order to increase the workability and flexibility of biofilm films based on different starches, various plasticizers, usually polyols have been widely used. Glycerol being one of the most preferred and most studied. Glycerol is a hydrophilic plasticizer and when added at the correct level with respect to the biopolymer content, can interfere with chain to chain hydrogen bonding and the water solubility of the biopolymer (protein/starch mixtures), a process generally used to improve the mechanical properties of biofilms [10].

APPLICATION OF STARCH

In the last many decades, scientists have employed the potential of starch to convert into films for various application in the field of packaging. However, satisfactory mechanical properties of starch converted into biodegradable film can only be achieved by jumbling with materials like lipids, hydro colloids, fillers, or active compounds. Several researchers have employed starch from various sources such as cereals, palms, tubers, roots most recently from psuedocereals for example *quinoa*, amaranth, achira etc., [11-15]. In some studies, it has been observed that some pseudo cereal grain has high starch and protein content [16]. The *Chenopodium* grain also consists high protein quality. It contains lysine, tryptophan, and isoleucine, not to mention that it is a potential source of dietary fiber, iron, calcium, sugars, polyphenols and other antioxidants [17]. The *Chenopodium* grain flour is a riveting available option to produce biodegradable composite films. *Chenopodium* (*Chenopodium album*) is an annual weed belonging to the Chenopodiaceae family; the *album*species of *Chenopodium* is grown widely in northern part of India and consumed as a food crop.

FILM PRODUCTION

Starch film is a thin continuous polymeric material. Various biopolymer film forming processes are available such as casting, spraying, extrusion and thermo-molding. The most commonly used process to produce films on a laboratory scale is casting, which is used to produce free films for testing. Casting method consists of drying drum for producing film with controlled thickness [18]. In this technique, with the help of drying for forming free standing starch cast on a non-adhesive surface, water or solvent is evaporated from the solution in order to form the film [19]. As a result of solvent evaporation, hydrogen bonds are formed and basic film structure is created. Environmental properties during the evaporation stage such as temperature and relative humidity could be used to control some of the film properties [20].

Table 1. Different types of starch, plasticizer and conditions to obtain starch film from casting

Reference	Type of starch	Plastisizer	Gelatization condition	Drying condition of film
Abdorreza et. al. 2011	Sago starch	Glycerol, Sorbitol	85°C, 30 min	40 °C, 20 h
Bertuzzi et al. 2007a, b	High-amylose corn starch	Glycerol	78–80 °C, 10 min	35 °C, 50 % RH
Araujo-Farr et al. 2010	Quinoa starch	Polyethylene glycol Glycerol	97 °C, 30 min	34–50 °C, 55 % RH
Pagella et al. 2002	High amylose maize starch	Glycerol, Sorbitol	90 °C, 2 h 90 °C,	23 °C, 60 % RH
Hu et al. 2009	Maize starch	Glycerol	85 °C, 3 h	45 °C for 8 h

Therefore, it come to the conclusion that the employment of starch films is a process which depends on several factors, such as the type of starch and plasticizer, which need to be optimized in order to produce films with adequate properties. Above Table summarizes the different casting methods with which to obtain starch films and highlights the differences between the methods used by different authors.

FILM CHARACTERIZATION

The various applications of the film are defined by a number of important film characteristics including WVP, tensile strength, elasticity, water or lipid solubility, and seal strength, peel strength etc.

These properties can be altered by addition of various components during film production procedure which may include plasticizers, cross-linking agent, antimicrobial agent, antioxidants and texture agent.

Chemical, thermal properties and color analysis

Thermal analysis is a branch of materials science where the properties of materials are studied as they change with temperature. Several methods are available that can be distinguished from one another by the property which is measured. The thermal properties of starch are determined with the aid of a differential scanning calorimeter equipped with a cooling system. The ash, protein, and moisture contents can be obtained using the standard AOAC methods [21].The lipid content can be calculated by the method of Bligh and Dyer [22]. These properties are necessary to determine as involved during the interactions taking place between starch, proteins, and lipids during drying of the filmogenic solution.

Visual aspect

The parameters of biodegradable films considered during visual aspect are: flexibility, ease of handling without rupturing, continuity and homogeneity [23]. Recent studies revealed that starch based film displays homogeneous and continuous surface without insoluble particles [24]. Canihua films of *Canihua cupi* and *Canihu*a *illpa-ini* reveals good handling and flexible properties which resembles its characterization to deals with more risk of fracture [25].

Moisture content

Moisture content is the quantity of water contained in a material, which can be films, paper sheet, wood etc. Water content is used in a wide range of scientific and technical areas, and is expressed as a ratio, which can range from 0 (completely dry) to the value of the materials' porosity at saturation. Moisture content of the film is necessary to define the physical property of the film. The standardize methodology for moisture content calculation used is ASTM D644-99 methodology [33]. The biodegradable film to have a good flexibility and resistant to microbial growth should have moisture content between 12-18%, below or above which will affect the film property [26].

Mechanical properties

The mechanical tests are conducted to analyze the strength of the film, it can be in the terms of tensile strength, seal strength, peel strength, elongation, and elastic modulus etc. These all are extensively important because packaging material must perform adequate mechanical strength to maintain their integrity during handling and storage. The tensile strength and elongation at break can be obtained using standardize methodology according to the ASTM D882-02 method [28].

Tensile strength of any film represents the resistance of the material to stand with the deformation and largely affected by the intra and intermolecular interactions.

Table 2: Mechanical properties of starch-based films

Reference	Film composition	Films conditioned	Elastic modulus (EM; MPa)	Tensile strength (TS; MPa)
Mali et al. 2006	Corn starch + 20 % glycerol	20 °C; 64 % RH	482 (86)	11 (3)
Dias et al. 2010	Rice starch + 20 % glycerol	25 °C; 58 % RH	532.8 (115.6)	10.9 (1.2)
Chung et al. 2010	Corn starch + 30 % glycerol	25 °C; 43 % RH	840 (61)	11.82 (0.57)
Mali et al. 2006	Cassava starch + 20 % glycerol	20 °C; 64 % RH	409 (30)	10 (3)
Hu et al. 2009	Rice starch + 30 % sorbitol	25 °C; 58 % RH	456.3 (81.0)	11.2 (1.0)

It can be seen that the higher molecular movement and their ability to form hydrogen bonding along the starch chain depends both on the types of plasticizer and the amount used. The main advantage of using plasticizers is that the film becomes more flexible, tensile strength (TS) is decreased and elongation at break (E %) increased [29].

Water vapour permeability

The water vapour permeability (WVP) of packaging film is widely determined by using the standard gravimetric method E96-95/ ASTM with modifications [30]. The WVP generally calculated at three different gradients of relative humidity (RH), low (2-33%), medium (33-65%), and high (65-90%), at 25ºC. Water vapor permeability defines the amount of water that can be transferred through the film under the certain atmospheric condition that depends upon relative humidity and temperature. WVP defines the rate which is measured as g/m^2/day i.e. amount of water in grams transferred per m^2 area in a day.

WVPR of a film is the major barrier property which is used to define the shelf life of packaged product. WVP plays an important role as a property of a packaging material as it avoids or decrease the moisture transfer to the minimum amount when compare to surrounding atmosphere.

Table 3: Water Vapor transfer rate of different starch based films

Reference	Film composition	Test conditions	WVP (g/m2/day)
Mali et al. 2006	Cassava starch + 20 % glycerol	25 °C; 0–75 %, fresh films	40.2
Dias et al. 2010	Rice starch + 20 % glycerol	25 °C; 0–75 %, fresh films	2.67
Jiménez et al. 2012	Corn starch/glycerol 1:0.25	25 °C; 0–75 %, fresh films	9.14
Mali et al. 2006	Corn starch + 20 % glycerol	25 °C; 0–75 %, fresh films	53.7
Dias et al. 2010	Rice starch + 30 % sorbitol	25 °C; 0–75 %, fresh films	3.03

Reportedly, the water vapor transfer rate of a starch based film increases as the moisture content and plasticizer concentration of starch films increase (Table 3).

Dias et al. (2010) observed that when the content of glycerol/sorbitol as plasticizer increased in the starch based biodegradable films, an increase in water vapor permeability was also observed [31]. It is also observed that starch based films developed with sorbitol as plasticizer had more moisture barrier than the films where glycerol is used as a plasticizer when tested in the same conditions (temperature and relative humidity gradient) which can be described with the relatively higher hydration ratio of glycerol than sorbitol as it is less hygroscopic in nature.

Color and Opacity

The concentration and the types of the plasticizers added proportionally influence the color of a film. Both color characteristic and opacity are directly linked to the visual aspects of the film. The morphology or the chemical structure related to molecular weight of the material affects the transparency and opacity of the polymer [32].

Color and opacity in various studies are analyzed on the basis of lightness, redness and yellowness. This is documented that the increasing glycerol level in the films lead to increase in reflection light on the film surface and recorded higher L-starch value. It indicated that the temperature has a significant influence in the structure of the films which in turn affect the cross-linking between amino groups in starch and reducing aldehyde and carbonyl groups leading to a tighten structure, thus results in yellowish color of the film [27].

Light Transmittance

Light transmittance indicates about light barrier property. This property is important to prevent lipid-oxidation induced by UV-light in food system and important parameter to evaluate the efficiency of packaging film for foods. Light transmittance of a film plays a very important role to prevent the transmission of harmful radiation through it and extent the shelf life of the packaged product by inhibiting the harmful radiation through it.

The transparency of the films is determined by measuring their light absorption at different wavelengths using a UV-Visible spectrophotometer according to the method described by Maran et al. (2013) [34].

Table 4:Light transmittance of canihua films of *Canihua cupi* film and *Canihuaillpa-inia* Film and light transmittance of air as compared with light transmission of a polyethylene film (PE)

Wavelength	*Canihua cupi* film (Tr %)	*Canihua illpa-inia* Film (Tr %)	Polyethylene (Tr %)	Reference
190	0	0	0	Lady M. Salas-Valeroa et. al
310	12	12	67	Lady M. Salas-Valeroa et. al.
430	19	19	72	Lady M. Salas-Valeroa et. al
550	26	24	76	Lady M. Salas-Valeroa et. al
670	33	29	80	Lady M. Salas-Valeroa et. al
790	45	41	90	Lady M. Salas-Valeroa et. al

According to the research work done for starch based biodegradable film, this Table demonstrates the Tr (%) in the films with the wavelength of 190 to 780 nm as compared with polyethylene (PE) films; air transmittance was the reference [35]. In comparison with polyethylene, the transmittance of C*anihua cupi* film and *Canihua illpa-inia* film reflect the presence of phenolic compound, which leads to the yellowish color of the film [36]. The organic substances which are having phenol group bear intramolecular hydrogen bonding to absorb UV light up to a certain extent. Hence, due to photo physical reaction they dissipate UV radiation in the form of thermal energy. Transmittance increase with glycerol and temperature i.e. higher amount of glycerol and temperature has greater transmittance [37-41].

CONCLUSION

Characterization and evaluation of starch based film is useful because it justifies the ability of starch to form biodegradable film with adequate properties. Starch as biodegradable packaging material can be socially and economically viable, and can be referred as an important tool to deal with existing challenges of petroleum based packaging materials and consequently resulting in enhanced shelf life, quality, safety, and security of foods. The starch based films also performed outstanding forming properties like air, moisture barrier, heat-sealing capacity, etc. Sometimes the addition of some additives in starch films is also required to produce a more ductile and flexible material, improving film handling. Corollary we need to improve functional properties of starch base films to match with petroleum polymer based films for which further studies are required.

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