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A REVIEW ON INTRODUCTION AND APPLICATIONS OF STARCH AND ITS BIODEGRADABLE POLYMERS

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Abstract

Biodegradable polymers play a very important role in plastic engineering by replacing non biodegradable, non renewable petrol based polymers. Starch is a renewable, biodegradable, low cost natural polymer with high availability. Natural polymers can be blended with synthetic polymers to improve their properties significantly. This article reviews advance in starch and starch based blends and presents their numerous potential applications. Therefore, this review helps to understand the importance and characteristics of starch and its biodegradable polymers (blends) by its various aspects such as structural properties and wide applications.

Keywords: Biodegradable polymers, starch, blends, application

Introduction

In the recent years, solid waste environmental pollution has increased tremendously because of the use of synthetic polymers. This is due to the fact that synthetic polymers such as polyethylene (PE) and polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS) persist for the long duration after their removal. The environmental friendly polymers are classified into two groups according to the raw materials: degradable synthetic polymers and renewable natural polymers (Yang *et al.,* 2004). Renewable natural polymers are starch, cellulose, soya and chitosan. Starch has been getting much more attention since 1970s (Griffin, 1994). Starch is the most attractive material among these renewable natural polymers; due to its low cost, availability and capability of high production from renewable resources (Guohua *et al.*, 2006; Tang *et al*., 2007; Yun *et al.*, 2008). Starch is renewable from carbondioxide (CO_2), water, sunlight by photosynthesis in plants (Teramoto *et al*., 2003). In this paper, structural properties of starch and applications of starch based biodegradable blends are discussed.

Structure and properties of starch

Numerous research articles have been published on the properties and chemistry of starch from different sources (Schwartz & Whistler, 2009; Whistler, Bemmiller, & Paschall, 1984). Starch is the reserve carbohydrate in plants tubes and seed endosperm plants (Otman, 2011) and occurs as granules in the cell in plastids, separated from the cytoplasm. The largest source of starch is corn and rice. Starch is a white soft amorphous powder and without sweetness. It is insoluble in water, alcohol and ether and it is non reducing carbohydrate (Jain *et al*., 2014). Starch granules are simple or compound, of varying size (2-150 µm), size distribution, and shape (Belitz & Grosch, 1999) (Figure 1). Starch is a mixture of two glucans, amylose and amylopectin. Most starches contain 10-20% water soluble amylose and 80-90 % water insoluble amylopectin depending on the source (Ramesh *et al.*, 1999). Waxy or glutinous starch from corn and other cereals contains little or no amylase, while a sugary mutant corn and some of the legumes contain amylose in greater abundance than amylopectin (Meyer, 1960). In addition to these glucans, small amounts of proteins and lipids are also present in starch (Belitz & Grosch, 1999).

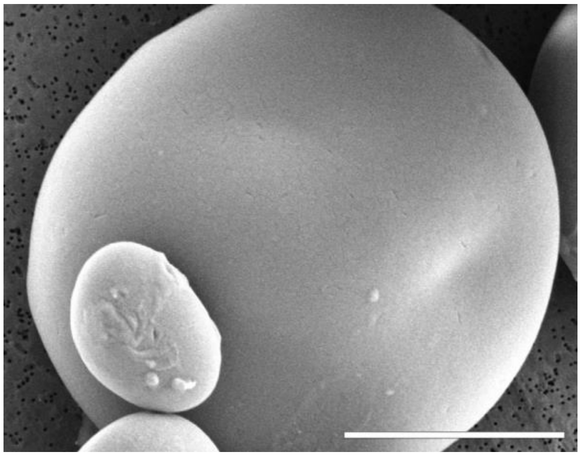

Fig. 1. SEM images of native wheat starch granules. The granules have a bimodal size distribution consisting of small and medium size granules. (scale bar = 10µm) (Source Glenn *et al.,* 2008)

Amylose is straight chain polysaccharides in which α-D-glucose units are joined 1-4. Chain lengths vary from 250 to 350 glucose units, and the long molecules appear to be coiled in α helix. Amylose is soluble in water (Wallace *et al*., 1981) but forms hydrated micelles. In such micelles the long chain is twisted into a helical coil. This structure is responsible for the blue color produced by iodine with starch (Manay & Shadaksharaswamy, 1997). The structure of amylase contributes to the gelling characteristics of cooked and cooled starches. The molecular structure of amylopectin is presented in Fig. 2.

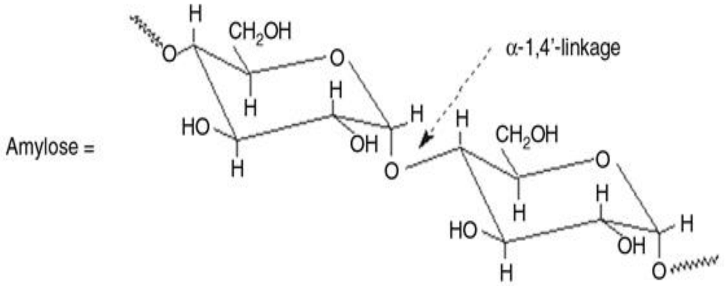

Fig. 2. Molecular structure of amylose (source Lu *et al.,* 2009)

Amylopectin also has a backbone of α- (1→4) linkages but, in addition, the molecule is branched through α- (1→6) linkages to the extent 4-5 percent. The length of the linear unit in amylopectin is about 20-25 percent glucose units. Amylopectin is responsible for the thickened properties of starch preparations but it does not contribute to gel formation (Manay & Shadaksharaswamy, 1997). The partial structure of amylopectin is presented in Fig. 3.

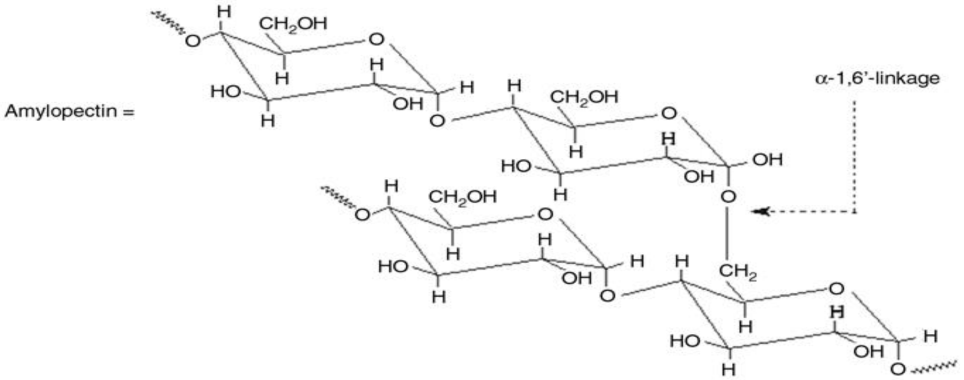

Fig. 3. Molecular structure of amylopectin (source Lu *et al.*, 2009)

On the basis of X-ray diffraction, starch granules possess semi-crystalline structure; with 70% amorphous and 30% crystalline in nature. The crystalline regions consist of mainly amylopectin whereas amorphous regions contain primarily amylase (Belitz & Grosch, 1999). The degree of crystallinity depends upon the moisture content of the starch. Dried starches show lower the degree of cystallinity (Belitz & Grosch, 1999).

Starch granules are hydrophilic in nature and consist of strong intermolecular interaction due to hydrogen bonding formed by the –OH group on the granule surface (Lu *et al.*, 2009). Chemical modifications are usually carried out to modify the starch. However, the properties of starch can be improved or modified by blending with different starches (Watershoot *et al.*, 2015).

Starch can be used in different ways:

- pure starch as thermoplastics
- its various derivatives
- starch through different reactive groups
- as blends by combining with other polymers
- as a copolymer with synthetic polymers

Starch is used as filler in blends to improve strength, minimize costs and increase the biodegradability of the materials.

Starch blends

Most of the biodegradable polymers obtained derived from renewable sources comprises hydrophilic character, fast degradation rate and poor mechanical properties. These limiting properties make the starch not suitable as a substituent for plastics. Due to these short comings, starch should be blended with synthetic polymers (Yu *et al.*, 2006). The first use of starch in plastics films was reported by Shulman and Howarth (Shulman & Howarth, 1964). The starch powder was used in making raincoats by the dispersion of starch powder in plastic films. The water vapor permeability of the film was increased by the starch, which allowed it to breathe and become more comfortable. Various starch based blends with different polyolefin were developed in 1970s and 1980s which are mostly non degradable (Yu *et al.*, 2006). So the many research has been carried out in the improvement of their biodegradability. The new low cost product with better performance can be developed by blending of biomaterials with synthetic polymers.

Blending approaches improved the mechanical properties of starch-films. Starch, lime, and citrus pectin were blended to characterize their ability to form strong self-supporting films with the use of different plasticizer such as glycerin, urea, and polyethylene glycol. They reported that thus prepared films possess strong high modulus, and among these plasticizers, glycerin showed best result (Coffin & Fishman, 1993). Blends of starch with synthetic biodegradable polymers have been widely studied (Lawton *et al.*, 1994; Wang *et al.*, 2003).

Verhoogt *et al.* prepared true blends of thermoplastic starch with poly (hydroxybutyrate-co-12% - hydroxyvalerate) where starch phase is deformed and are brittle in nature. Thermal analysis showed there is no interaction between the two polymers (Verhoogt *et al.* 1995).

Many researchers have been worked out in the field of starch nanocomposites. The properties of TPS can be improved by making its composites. Park *et al.* (2002, 2003) prepared the gelatinized starch/montmorillonite (MMT) clay nanocomposites using both naturally occurring sodium MMT and number of alkyl ammonium modified clays and investigated their properties. An intercalated structure for the starch/sodium MMT was observed whereas, either unchanged or agglomerated structure appeared by modified clays (Park *et al.*, 2002; Park *et al.*, 2003). Unmodified sod. MMT/starch nanocomposites displayed the greatest increase in modulus of all the clays used was reported by Park *et al.* (2002**).** The barrier properties of composites were found increased with the function of clay content was mentioned by Park *et al.* (2003). There was increment in dynamic elastic moduli and tensile strength due to dispersion of the clay platelets in the starch matrix (Figure 4). The water vapor transmission rate was significantly lowered by the platelets with generating a twist pathway through the starch matrix (Chiou *et al.*, 2005). Furthermore, the dispersed platelets increased surface area for starch/nanoclay interactions that, in turn, improve the mechanical properties of the starch composites.

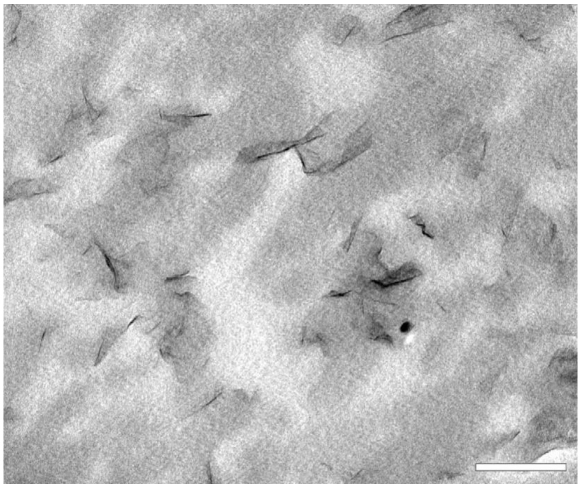

Fig. 4. TEM micrograph of starch/clay composites. The clay particles appear as dark lines dispersed throughout the starch matrix. (scale bar = 200 nm) (Source Glenn *et al.*, 2014)

Fischer & Fischer (2001, 2003) investigated various experimental pathways, including the dispersion of Na^+ MMT clay in water, followed by blending in an extruder at a temperature of 85-105 °C with a premixed powder of potato starch, glycerol and water. The prepared materials were found exfoliated and showed less hydrophilicity, and enhanced stiffness, strength and toughness.

Huang *et al.* (2004) reported melt extrusion process to prepare MMT-reinforced glycerol/plasticized thermoplastic starch. Well dispersion of MMT platelets were seen in Scanning electron micrograph. Fourier transform infrared (FTIR) spectra indicated cooperation existed between MMT and starch molecules as well as existence of hydrogen bonds between the hydroxyl groups of MMT and the hydroxyl groups of starch molecules. The nanocomposites of starch showed significant improvements of their mechanical and thermal properties.

Otman, 2011 studied the thermal properties of polyvinyl alcohol (PVOH)/corn starch composites and found that the thermal stability of polyvinyl alcohol was suppressed and the PVOH-corn starch system were miscible (Otman *et al.*, 2011).
Liu, 2013 prepared noble plasticized starch using diethanolamine as plasticizer and then fabricated polypropylene (PP)/starch blends using maleic anhydride grafted polypropylene as the compatibilizers. They reported enhanced thermal and mechanical properties due to improved interfacial adhesion (Liu *et al.*, 2013).

Lukasiewicz, 2014 investigated the ternary system of polyaniline (PAN)/starch/hydrocolloid and observed that polysaccharide matrices are appropriate for making the oxidative polymerization reaction at ambient temperature (Lukasiewicz *et al.*, 2014).

Most popular packaging and disposing materials are thermoplastic starch (TPS) due to its properties, availability and cost effectiveness. Trovatti *et al.* (2014) prepared the system of TPS with oxidized natural rubber by extrusion method and reported expected improvements having excellent water resistance and decreased fragility (Trovatti *et al.,* 2014).

Bin-Dahman *et al.* (2015) studied the compatibility of poly (acrylic acid) (PAA) and corn starch by various methods. The blends of PAA/corn starch with glycerol as a plasticizer prepared by solution casting technique and found molecular level interaction between the blend components and their partial miscibility and amorphous structure was confirmed by differential scanning calorimetry (DSC). Thermogravimetric analysis (TGA) revealed the more thermal stability of blends than individual polymer (Bin-Dahman *et al.,* 2015).

Application of starch and starch based biodegradable blends

The industrial utilization of native starch is limited to inherent high rate of retrogradation, insolubility in water, fluctuation in viscosity during thermal processing (Ashogbon & Akinatayo, 2014). This deficiency of native starches is mitigated by physical and chemical modification. The chemical and physical modifications of starch are more expensive and repeatedly utilize treatments with hazardous synthetic chemicals for human and animal health (Sanstelia & Zeeman, 2011). Currently, people are avoiding the chemical applications in natural food products (Zhang *et al,* 2011). Ashogbon, 2014 prepared blends of different starch such as bambara groundnut starch, cassava starch, cocoyam starch and wheat starch in different proportions and evaluated their physicochemical properties. They reported that blending of starch from different botanical sources improved their properties (Ashogbon & Akinatayo, 2014).

It was reported that starch is an important thickening and binding agent and is used to a large extent in pudding, soups, sauces, salad dressings, diet food preparations for infants, pastry filling, mayonnaise etc (Belitz & Grosch, 1999). There are many applications of starch based biodegradable blends in various fields such as in food industry, in agriculture, tissue engineering and drug delivery systems.

Most of the packaging materials make up the largest sector (29.5%) of plastic waste in MSW (EPA, 2010). At present, packaging plastic has become an integral part of the worldwide market (WPO, 2008). Starch based biodegradable polymer are used in food packaging and in edible wrapping or tubing. Edible films of starch are colorless, odorless, tasteless, non toxic and biodegradable. They show very low permeability to oxygen at low relative humidity

(Phan The *et al.,* 2009) and are suggested their uses in food product protection to improve quality and shelf life (Flores *et al.,* 2007).

The starch based biodegradable polymer can be an effective alternative for food packaging to resolve the problem of environmental pollution, health and disposal problems (Ozdemir *et al.,* 2004). Starch/clay nanocomposites possess better mechanical property and lower migration of polymers and additives are developed on food packaging materials (Avella *et al.,* 2005).

Starch based biodegradable polymers have been widely investigated in bone tissue engineering scaffold (Gomes *et al.,* 2003). They are also used in drug delivery system in the form of microsphere or hydrogel (Balmayor *et al.,* 2008; Reis *et al.,* 2008). So there is no need for surgical removal of the devices after their drug depletion. Starch/gelatin blend microparticles are proposed to use in controlled release delivery carrier for water soluble drugs (Phromsopha *et al.,* 2014).

Starch based polymers are biodegradable and have three major applications in agriculture (Dilara *et al.,* 2000) such as the covering of green house, mulch films and fertilizers. Film made of starch blends was the first film containing renewable content tested as agricultural mulch (Otey, Mark, Mehltretter, & Russel, 1974). Currently, poly lactic acid, a polymer derived from starch fermentation was fabricated with fiber to make composite films for mulching application in agriculture. The starch-based controlled release devices have been prepared to manage parasitic mites in honeybee colonies (Glenn *et al.,* 2006).

Biodegradable polymers play a very important role in plastic engineering by replacing non biodegradable, non renewable petrol based polymers. Starch/poly (lactic acid) blends can potentially be used for tooth brush handles, flower pots and cups (Kovacs *et al*., 2011)

The importance of starch-based biodegradable is increased in the condition of the greening of industrial chemistry. Therefore, researchers are interested to develop the biodegradable polymers. The demand of biodegradable polymer industry has been increased significantly, so, there is bright prospect mainly due to environmental benefits of using renewable resource feedstock sources.

Conclusion

Starch is a natural polymer which is renewable from carbon dioxide, water and sunlight by the photosynthesis. It is biodegradable, physically or chemically modified and inexpensive. Some of the prospects of starch have been highlighted and looking potential direction for future research. Therefore, different approaches or strategies are needed to develop starch based biodegradable polymers industry for enhancing their biocompatibility, bio-degradability and physical properties for their diverse applications.

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