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Starch based polymeric composite for food packaging applications

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Abstract
The starch-based polymer is an incredible substitute to petroleum-based plastics.It has an extraordinary possibility to form an edible, biodegradable film by blending in with various antimicrobial polymers, nanoparticles, and other antimicrobial blenders, for instance, essential oils from phenolic extracts. Characteristics, preparation methods , physicochemical properties, different modification methods for improvement of starch film properties are studid in this paper. This study surveys the sensibility of starch for food packaging.
Keywords: Starch; Antimicrobial activity; Antioxidant; Mechanical properties; Optical properties; Barrier Properties

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I. INTRODUCTION

For packaging material, plastics have been utilized generally quite a while back (Nattakan et al). Plastic wastes cause environmental pollution since it can't biodegraded by microorganism. In 2015 plastics created around 322 million tons around the world, which is a 3.5% expansion when contrasted with 2014.In (2014–15) 8.3 million tons of plastics were delivered in India (European Bioplasticsa (2016)). Now, about 99% plastic materials are manufactured from petroleum based (non-renewable) resources. To diminish natural contamination just as because of the vulnerability of petroleum supply as of late there is a huge advancement in the improvement of biodegradable products dependent on farming materials. The bio-based polymers those can be utilized for the preparation of films for food packaging applications are polysaccharides, (starch, alginate, cellulose, Chitosan, carrageen),proteins(whey protein, corn Zein) and their subordinates (Gago et al.).The starch films are brittle and hard to process; plasticizers are generally added to the film forming solutions to get rid from film brittleness. (Carmen et al).To improve starch based film properties numerous researchers examined that various fillers can be utilized as reinforcement to increase the tensile strength and elastic modulus and diminishes their elongation capacity(Avérous et al.).Normal fillers are powder, calcium carbonate and barium sulfate. Other mineral fillers utilized are wollastonite and mica. Some mineral filler is surface treated to improve their characteristics performance (Sudhin et al.). Mostly Silanes, glycols, and stearates fillers are used for improvement in processing and dispersion. The aim of this research is to examine the connection between the physical-mechanical reaction and the structure of starches with possible applications in the production of food packaging.

II. STARCH: SOURCE, STRUCTURE AND PROPERTIES

2.1.Source of starch:

There are five main raw materials for starch such as: maize, wheat, potato, sweet potato, and cassava as shown in figure 1. In Europe wheat starch extraction, mainly occurs. But wheat starch and cassava starch are cultivated in Brazil. Sweet potato starch production mainly occurs in Asia. Mostly cornstarch is used for production of biodegradable polymer film because 99% starch production occurs from corn. Corn starch mostly happens in the United States, China, and Brazil. Brazil likewise a huge cassava producer, as indicated by the Food and Agriculture Organization. In excess of 100 nations created Cassava with a complete generation of 273 million tons. Brazil is in fourth position for generation of Cassava.Annual creation of Cassava in Brazil is 23 million tons. Nigeria, Thailand, and Indonesia, whose creations have arrived at 55, 30, and 23 million tons, individually. As indicated by Food and Agriculture Organization information after corn, cassava gives most prominent creation of starch.

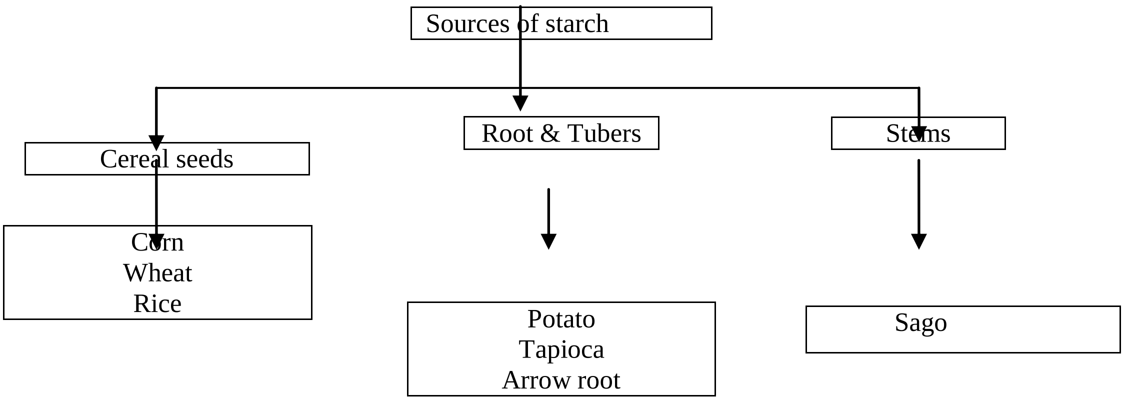

Figure1. Sources of starch

2.2 Structure of starch based on the source

Starch is a semicrystalline polymer comprise of two types of molecules, amylose (20-30%) and amylopectin (70-80%).In amylase glucose molecules connected by a-1,4 glycosidic linkages with the ring oxygen atoms all on a similar side. Amylopectin is a huge extended polymer with α-1,4 linkage which provides support and α-1,6 bridges at the bracnching point. Figure (2) displays the amylose and amylopectin molecules. The proportion of amylose to amylopectin contrasts from starch to starch. It relies on the botanic beginning of starch (Darker and Poon). Hence they show various properties Table (1). The proportion of amylose/amylopectin relies upon the source and age of starch Table (2).Generally, native starches are in granules form and amylose(crystalline) and amylopectin (non-crystalline) of native starches are arranged in alternating layers. Based on the starch source crystalline structure types are formed, for example, An (oats), B (tubers), C (a blend of An and B types), and V (retrograded starch). During the film formation process, a high water content and high heat cause the starch granules to swell and, in this sense, the gelatinization of starch is caused by the interruption of the crystalline association. So to make successful commercial application of starch, gelatinization of starch and the complete dispersion of water are necessary since the water acts as a plasticizing agent(Copeland et al.).

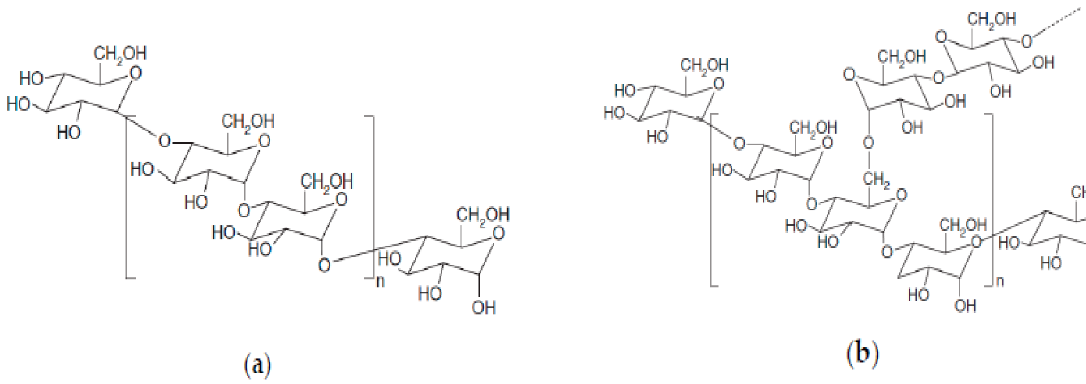

Figure 2. (a)Amylose (b) Amylopectin structures

Table 1. Properties of Amylose & Amylopectin

Properties	**Amylose**	**Amylopectin**
Shape	linear	Branched
Linkage	α-1,4	α-1,4 and α-1,6

Table 2. Characteristics of Amylose and Amylopectin.

Starch type	**Sources**	**Shape**	**Size (mm)**	**Amylose Percentage(%)**	**Amylopectin percentage(%)**
Wheat	Cereal	Round, lenticular	5–35	25	75

Potato	Tuber	spherical	10–100	20–30	80–70
Tapioca	Root	Oval	3–28	17	83
High-amylose corn	Cereal	Polygonal	---	55–70	45–30
Pea	Legume	Oval		25–40	75–60
Rice	Cereal	Polygonal	2–10	19	81

III. FILM PREPARATION: WET & DRY PROCESS

There are various methods reported in the literature for preparing of starch, for example, solution casting, injection molding, extrusion, and compression molding (Shanks and Kong, 2012). Basically, all these techniques divided in to two groups :A)Wet Process B)Dry Process. The thing that matters is the amount of required water for the procedure. The wet procedure includes a lot of water, and is therefore called the wet-technique. The next one required a lower amount of water as compaire to the wet procedure (Jiménez, Fabra, Talens and Chiralt, 2012) and for this reason it is called dry process.

3.1 Wet Process

In wet procedure film is generally formed by two methods, such as, I) Dispersion or ii) emulsion. The wet procedure is otherwise called solvent casting method. This method is most commonly used to obtain edible films. In the solvent casting technique, the polymeric solution is poured on to a flat surface such as a circular plate/ dish (Bertuzzi et al.) Or a Teflon® plate (Jiménez et al.) to get a uniform film.

Numerous scientists revealed that starch films acquired by casting (Bertuzzi et al. ;Dias et al. ; Talja et al.). The total procedure divided into four main steps, for example, gelatinization and dispersion, homogenization, casting, and drying.

The first stage is the gelatinization. Depending upon the type of starch & plasticizers content gelatinization takes place at different temperatures. Starch and plasticizer interconnected by hydrogen bonding. During processing, by increasing the process temperature causes an increase in matrix mobility, as a result, there is a decrease of viscosity. It results in smoothening of starch processing. (S. L. Rosen; M.G.A. Vieira ; A.L.M. Smits). For lipids containing films, emulsions are obtained by using rotor-stator homogenizer. Garcia et al. reported that, they have utilized Fischer Scientific fast homogenizer to acquire emulsions of beeswax–pea starch and sunflower oil–corn starch. Jimenez et al. also used a homogenizer to homogenize a mixture of fatty acid–glycerol–corn starch. Ahmed et al. prepared cassava starch-based film by casting method. They arranged film with 5gm of cassava in 100ml of refined water and plasticizers in concentrations of 0.30, 0.45, and 0.60 g/g dry starch. They also used stirrer for homogenize mixing of components. Chinma et al.prepared an edible film utilizing different concentrations of cassava starch and soy-protein. They heated the film-forming suspension in a flask and consistently blended in with a glass bar to get a film-forming solution. After homogenization film-forming solution was cooled and cast onto a level bar plate for the setting. At the point when set, they are permitted to dry before expelling the film from the plate (Chinma et al). Various surfaces can be used,e.g. hardened steel (Hu et al.) or glass (Flores et al.), to accomplish a better than average appearance in starch films. The gelatinization condition and drying time change for different starch-based film as appeared in table 3.

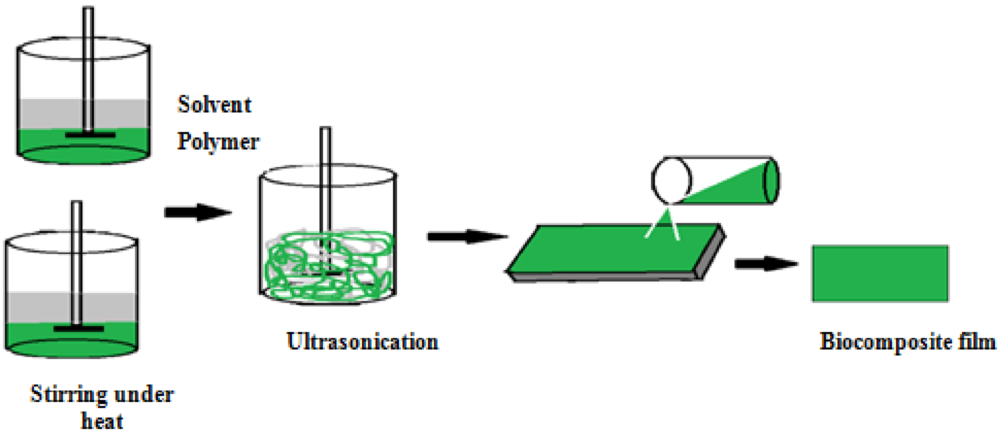

Fig.2.Flow diagram of starch film production by casting method.

Table 3. Gelatinization conditions and drying conditions of various types of starch basedfilms.

Starch source	**Drying conditions**	**Gelatinization conditions**	**References**
Sago	40 ^{0}C, 20 hr	30 min,85 ^{0}C	Abdorreza et al.
Pea	Room temp,40 hr	15 min, Boiling temp	Han et al.
Quinoa	34–50 ^{0}C, 55 % RH, for different times	30 min, 97 ^{0}C	Araujo-Farro et al.
Cassava	30 ^{0}C, 24 hr	1 min, 70 ^{0}C	Bergo et al.
Corn	60 ^{0}C, 8 hr	Alkaline medium	García et al.

3.2 Dry Process

Processing of edible film using industrial process in some cases is not possible. For the production of starch biocomposite the starch is used in granular form and sometimes melt blended in polymer matrix. As casting techniques is preferred for biocomposite production but for large scale production dry process should be considered.

For modification of synthetic polymer extrusion process generally used but now this process is also used for starch modification . A co-rotating, twin-screw extruder is used to prepare the biocomposite starch. During the extrusion, the raw materials are forced through a die to form products.Some researchers also used two stage sheet or film extrusion techniques (Fishman, Coffin et al. 2000; Matzinos, Tserki et al. 2002). Manoel et al. The work presented a typical example of film extrusion. They combined the materials such as waxy starch, glycerol and water by a twin screw extruder .The composites were granulated and then exruded as a film using a single screw extruder. Shirai et al.prepaired biodegradable flexible films of starch and poly(lactic acid) plasticized with adipate or citrate esters by blown film extrusion method.They produced pellets in two stages by twin screw extruder. Finally film is produced in blown film extrusion method . under these conditions; such as: a screw diameter of 25 mm, a screw speed of 35 rpm and a temperature profile of 100/150/150/100 ◦C. Calleja et al. prepared potato starch film in two different method such as i)injection molding ii)compression molding and observed the effect of processing method on the properties of potato starch film. After processing in above two mentioned methods, the samples were subjected to ambient atmosphere and stored about 4 weeks for investigation. They reported that sample formed from injection molding process showed better mechanical properties than the samples formed from compression molding. Pelissari et al.reported how extrusion parameter affects physical properties of starch film.they prepared starch film blended with chitosen and glycerol by extrusion method. To get good mechanical and barrier properties they have used 77% of starch,5 % chitosan,18% glycerol. 0.5% concentration of oregano essential oil (OEO) also added in order to inhibit to inhibit Escherichia coli, Staphylococcus aureus, Salmonella enteritides and Bacillus cereus . At first stage the mixtures were fed in to a extruder with a temperature profile of 120 ⁄ 120 ⁄ 120 ⁄ 110^{0}C and screw speed of 35 r.p.m.). At last stage, according to the experimental design die temperature and screw speed were varied. They concluded from the experiment that with the increase in screw speed there was increase in BUR and and water vapour permeability (WVP) aand decrease in tensile strength. They also reported that extrusion conditions also affects the absorption isotherm of films . T. Tábi, J. G. Kovács reported the effect of injection molding & extrusion molding processing techniques on properties of starch film.They used maize starch (the ratio of amylase-amylopectin was 25:75),glycerol & distilled water.TPS was processed in two stage.first in extrusion process then injection molding process.In extrusion process they processed the mixture to produce pellets then that pellets are injection molded to produce TPS film. They studid the effects of injection moulding parameters and storing methods on properties of TPS film. They found that aeging is faster for samples those were stored in ambient conditions .The holding pressure had effect on the tensile modulus of the specimens.The results of Ming Li researchers showed that amylase content affects the processibility and mechanical performance of corn starch. For extrusion processing of high-amylose content starches, high processing temperature is required as processibility decreases with increasing high amylase content. In an examination, for the proper blending of starch with alcohol, Gilfillan et al. used thermal extrusion process so as to improve the film properties. During processing improvement in starch film properties occurred due to the formation of hydrogen bond or it may be due to formation of carboxylate group by the oxidation of the glucose unit of starch during extrusion stage. Qianqian et al. have investigated effects of glycerol concentration and extrusion processing process on the properties of oxidized and acetylated corn starch-based films. They reported that extruded film exhibit poor tensile strength, high water vapour permeability and higher oil permeability than as compared to the non extruded film. In untreated film by increasing glycerol content exhibited same properties like treated film. Extrusion process conditions affects starch film morphology (Seligra et al). They investigated the effect of screw speed on extruded thermoplastic starch materials. Starch processed at 80rpm screw speed caused fully gelatinized where as at screw speed at 40rpm and 120 rpm caused broken starch grain. But after pressing the thread made at 120 rpm caused no broken grain starch. 80 rpm screw speed is suitable to obtain homogenous thermoplastic starch material with a low tensile strength and high starch retrogradation.But starch film with 120 rpm screw speed resulted high tensile strength,greater modulus and slower starch retrogradation. Research has showed that how starch resin moisture level affect the injection moulding processability(Jordan M. Ellingson). TPS/HDPE blends containing 0.6% to 1.4% moisture level voids percent increased by 300-350% (void percentage measured by cross section area) where as in unblended thermoplastic starch , for moisture levels 0.4% to 1.4% void percent increased by 150%.moisture of TPS didnot affect tensile strength of TPS/HDPE blend but it affected tensile strength of TPS/PP blend.

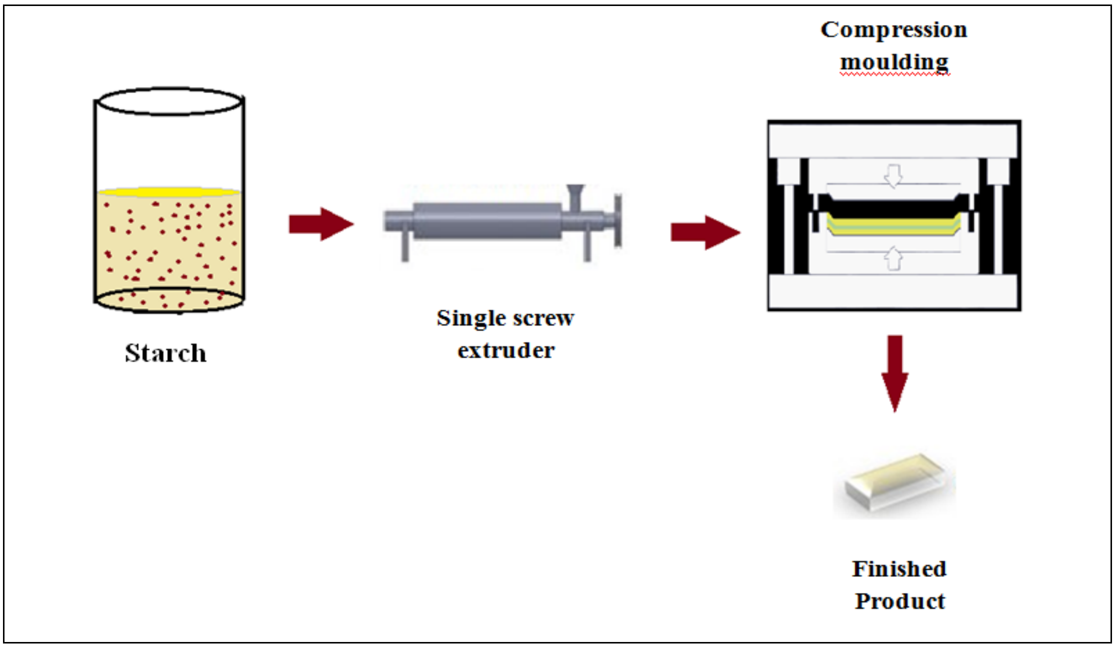

Fig.2.Flow diagram of extrusion process for starch film production.

Table 1:
The list of woks has been carried out on starch based biopolymers.

Starch blend type	**Film processing method**	**Methodology**	**Characterization techniques**	**Observation**	**Reference**
Blending of starch with glycerol and water.	Injection & Extrusion moulding method.	starch:glycerol:water ratio was 70:16:14 and glycerol-mono-stearate was added as a lubricant.	SEM	The starch film processed by two methods. First extrusion then injection moulding method. According to the SEM images , after extrusion some starch granules were still remained in TPS pellets but at the second stage that is when the pellets were injection moulded remained starch granules are disappeared.They also studid the effect of various injection moulding parameters on starch film. These films have shown that ageing occurs more quickly in TPS film stored in ambient conditions than 50% relative humidity .The tensile stress didn't affect by holding pressure but tensile modulus affected by it.	T. Tábi, J. G.
Blending of Cassava starch with glycerol and clay nanoparticles .	Solution casting method	The film was developed and the influence of the glycerol and clay nanoparticles on the tensile and barrier properties and on the glass transition temperature was investigated.	FTIR, XRD, SEM	The obtained mixture has shown that the percentage of elongation(%E) is decreased for the films prepared without the addition of clay nanoparticles and has increased the addition of the glycerol content, but the films prepared with the addition of the content of nanoparticles (0.10 g/100 g) showed a high percentage of Elongation(%E).they also observed that addition of clay nanoparticles increase barrier properties of film. They also studied that Tg decreases when the glycerol content increases and increase of nanoparticle content has no significant effect on Tg.	Souza et al.
Corn starch blend with PLA and plasticized with adipate and esters.	Extrusion method	PLA/Starch based polymers were developed. The film was prepared in two phases. In the first phase, TPS 30% (w/w) and PLA 70% (w/w) were extruded in a twin-screw extruder and formed pellets and in the second stage that pellets were injection moulded to form film. The effects of the plasticizer on the film's properties have been studid.	SEM, TENSILE TEST, APARENT OPACITY	In the mechanical test of starch/PLA blend plasticized with adipate and esters resulted that plasticizers do not have a significant effect on the mechanical properties of the film. Films prepared with adipate esters showed high elongation (120–148%), low tensile strength (0.6–0.9 MPa), and a low Young's modulus (1.6–3.8 MPa) as compared to films made with citrate esters. Opacity test of Starch/PLA film concluded that opacity of film decreased by increasing the concentration of adipate or ester plasticizers. Structural analysis of Starch/PLA blend was conducted by SEM. From SEM it was observed that blend containing no phase separation.	Shirai et al.
Blending of starch and chitosan	Extrusion method	The film was prepared by the extrusion method. The percentage of maize starch: glycerol:water	SEM, FTRI, XRD, TGA, TENSILE TEST	From the FTIR spectra it was showed that spectra of TPS/TPC film very much similar to pure TPS film. TPS/TPC film showed same XRD diffraction pattern like pure TPS film. From the SEM experiment it was observed that TPS/TPC film surface have	Mendes et al.

		was (60:24:15.% by weight).		some surface cracks. Mass loss of TPS/TPC film was observed in the temperature ranges of 25–160◦C, 160–500◦C and 500–600◦C, as detected by TGA. The tensile test reported that the addition of TPC reduced the tensile strength of starch/chitosan composite.	
Cassava starch/Soya protein edible film	Solution casting method	Cassava starch/Soya protein edible film was developed using solution casting method. The mixture was prepared by taking different proportion of Cassava Starch, Soya protein such as, (100: 0 %; 90: 10 %; 80: 20 %; 70: 30 %; 60;40 % and 50: 50 %)	Tensile test, WVP test.	The impacts of relative humidity and temperature on the mechanical properties and water vapor permeability of cassava starch/soy protein films were observed. In the WVP test it was found that with the increase of temperature and relative humidity, WVP of the cassava starch/soy protein film has increased. The tensile test showed that with increasing of temperature at a constant relative humidity, the tensile strength of the Starch/Soya protein film increased, but decreased as relative humidity increased.	Chinma et al.
Biodegradable films from Rice and Pea starch.	Solvent Casting method	The film was developed and the effect of relative humidity and plasticizer content on the physical & mechanical properties of the rice and pea starch film was studid.	Photomicrography test, Tensile test, WVP test, Oxygen permeability test.	From photomicrography it was seen that plasticizer has a strong effect on the structure of rice and pea starch films. It prevents accumulation of amylase and amylopectin as a result homogeneous structure obtained. Plasticized RS and PS film held 15.4% to 15.6% more water than unplasticized film after conditioning at 51% RH. Glycerol has good water holding capacity, so it act as a good plasticizer for RS and PS film. Humidity has very less effect on RH film. Additionof plasticizer caused decreased of tensile strength and elongation in both RS and PS. Above 40%RH oxygen permeability of both the film was in the range(1.2 to 1.4 cm3 µm/m2/d/kPa).	Ghadeer F Mehyar, Jung H. Han.
Cassava starch	Casting	The film was prepared by the casting method and various plasticizers effects on the thermal, physical, mechanical, and structural properties of the cassava-starch-based films were studid.	WA, DSC, TGA, SEM, FTIR, XRD, Tensile test	From the water absorption test it emerged that the increase in plasticizer concentration causes a greater water absorption in the cassava starch film. The DSC study revealed that plasticizer addition builds the initial temperature and the maximum temperature of the starch films contrasted with the control film.So, sealing strength of cassava starch film was increased.TGA resulted that increasing concentration of plasticizer about 40 to 60% thermal resistance of the film decreases. From SEM study it resulted that addition of 45% plasticizer caused more homogeneous structure in the film as compared to addition of 30% plasticizer. FTIR study showed that fructose and urea formed strong interaction with cassava starch as compared to other plasticizers such as TEA and TEG. XRD results indicated that the crystalline structure of the native starch has been destroyed.	Edhirej et al.

				Types and concentration of plasticizers did not affect crystallinity of cassava starch film. From the tensile test they found that the increase in plasticizer concentration caused a decrease in the film's tensile strength. Fructose plasticized film resulted good tensile strength as compared to other plasticized film.	
corn starch with formamide and urea plasticizer/natural fiber	Melt mixing	Plasticizer mixed with cornstarch and natural fiber using a high speed mixture. Thus, that mixture was reintroduced in to a single screw extruder to form a starch film sheet. The effect of fiber concentration on the mechanical and thermal properties of the film has been studid.	SEM , Tensile test, XRD, Water absorption test, TG	The mechanical test showed how the concentration of fiber and water absorption influenced the mechanical characteristics of the starch film. By increasing the fiber concentration to 20%,the tensile strength has tripled up to 15.16 Mpa. but when the water content increased, then fibre had no effect on the tensile strength of the film. XRD study informed that below 20% fibre concentration matrix restrained the retrogation.Improvement in thermal stability of film was confirmed by TG analysis. From the water absorption test ,they found that increment of fibre concentration caused less water absorption.	Xiaofei et al.
Tapioca starch/glycerol/ micro crystalline cellulose	Casting	Starch with crystalline cellulose prepared with the solvent casting method was observed and the effect of micro crystalline cellulose on the physical, mechanical, thermal and barrier properties of the film was observed.	SEM, TENSILE, WVP, DSC	It was found that by adding 3wt% of micro crystalline cellulose(MCC) fiber resulted good mechanical and barrier properties.SEM results showed that addition of 3wt% of MCC in to starch caused uniform surface finish whereas addition of 10wt% MCC caused agglomeration in starch matrix.Tg & Tm of film increased by addition of 3wt % MCC. I The TPS/MCC films produced in this study contained improved properties have the potential to be used for the application of food packaging.	Othman et al.(2019)

Starch/clay nanocomposite	Polymer melt processing technique	The clay nanoparticle has been incorporated in to various starch based materials using polymer melt processing techniques. The Structural & mechanical properties of nanocomposite films were studid.	NMR TENSILE CONTACT TEST	NMR analysis resulted that clay nanoparticles uniformly dispersed in the starch matrix.As a result, due to the reinforcing effect of the clay nanoparticle modulus and the tensile strength of the starch/clay film, it has also been increased. To confirm the samples with the current regulations and the European directives on biodegradable materials, a contact test was performed for the starch/clay film sample.	Maurizio et al.(2004)

Cassava starch	Blown film extrusion	The starch mixture was prepared using 1kg of cassava starch, 2kg of liquid polyvinyl alcohol , 100g of talc powder, and 100g of urea. After the preparation of the mixture, that mixture was extruded with a blown-film extruder to produce a biodegradable film. Tensile test and biodegrdibilty test were performed.	TENSILE, BIODEGRA DATION	Tensile test of starch film was carried out and it was compaired with polyethylene and paper .it was confirmed that starch film showed highest tensile strength as compaird to polyethylene and paper. biodegradability test reported that starch film contain organic matter than which is less than paper but more than polyethylene.Hence it is concluded that cassava starch film can be biodegradable and it can be used as a substitute of conventional material.	S. L. Ezeoha, J. N. Ezenwanne(2013)

IV. PHYSICAL PROPERTIES OF STARCH FILMS

4.1 Hydrophilic Nature of Starch-Based Materials

Hydrophilic nature of TPS, limiting its applications. Water absorbance of starch as a function of the atmospheric relative humidity (RH). Starch can absorb significant quantity of water. Modification of starch morphology and internal interaction should be possible by water molecules along these lines its mechanical properties and glass transition temperature rely upon the water content (WC). Furthermore, plasticizers additionally diminish the intermolecular forces in polymer and improve ductility of the film as shown in figure 3.

The Optical and barrier properties of the starch film can be impacted by the composition (starch, glycerol, tapioca, agar) of the starch film(Prakash Maran et al.).According to authors, there is a relationship between solubility capacity of starch film with respect to glycerol concentration and surfactants. Solubility capacity of starch films directly proportional to glycerol concentrations and inversely proportional to surfactants (span80) concentration, this is because of the hydrophobic character of starch film. By reduction of O-H bond the hydrophilicity decreases,as a result solubility of film also decreases.

Flores et al. explained the effects of processing techniques on physical properties of tapioca starch films containing sorbate. For all processing techniques, the solubility of the films applied at 24 hours was the same (≈22%) but increased when sorbate was present in the films. Antimicrobial presence causes this differences. Furthermore, the increase in the addition of essential oils caused decrease of solubility of the film .

Kim et al. explained how solubilityof starch film depends upon pullan percentage in composite films based on tapioca starch and pullulan. According to authors, solubility reached higher than 40% at 60 s when the pullulan was continuously leached from the pullulan film, but the starch film displayed very minor solubility (about 3% after 60 s).

Kuorwel et al. revealed various properties of TPS films after introduction into different compositions of water mixtures and various level of RH by Conditioning the starch films at different relative humidity(RH) that brought about a recognizable absorption of moisture, especially at high RH. The authors recommend that the higher moisture absorption at RH is because of the great connection between water, starch and the plasticizer, which causes weakening of the film. Although the low-moisture absorption of the starch film at low RH may be because of weak interactions between the starch matrix and water molecules.

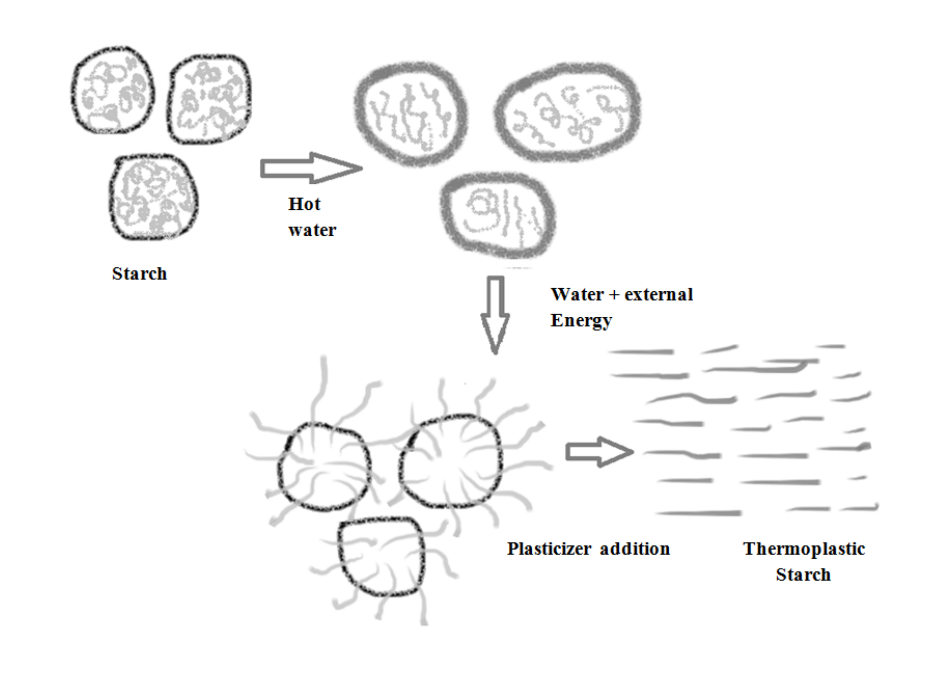

Fig.3. Hydrophilic nature of starch molecules

4.2 Mechanical Properties

Starch films are strong materials with a stress at break of up to 90 MPa (Lafargue et al., 2007). Mechanical stability of starch decreases with very high humidity.At 60% relative humidity stress at break decreases up to 40MPa and at high humidity stress at break value decrease up to 10MPa (Koch et al., 2010).The factors that influence tensile properties of starch are are temperature, air humidity, plasticizers, the type of starch, additives, Tg and crystalline, the last being firmly identified with the amylose: amylopectin relationship

Starch having two primary parts, for example, amylose and amylopectin, with various mechanical properties because film of pure amylose is stronger than pure amylopectin films . addition of plasticizers affects the crystallinity of amylase and amylopectin films. Causes of glycerol additions, increased crystallinity with increased air humidity. The presence of a plasticizer and longer drying time favor the mobility of the chain by advancing in the improvement of the crystallites. (Menzel et al., 2015).

Ahmed et al. reported that the tensile strength of the starch film has diminished totally with the increase of plasticizer content.With addition of 30% glycerol the tensile stress was 2.68MPa . But when 60% tri-ethylene glycol(TEA) added to the film ,it showed that films containing 60% of TEA indicated lower tensile stress values because of their hygroscopic nature which will, in general, give extra water in the film matrix.PVA/Starch plasticized with epoxidized soyabean oil delivered a tensile strength estimation of 6.7MPa and 13.6MPa for 20% and 30% PVA contrasted with PVA/starch plasticized with sorbitol. The ether bond in the molecular structure of ESO has expanded the dipole bond between the polymer chains. Thus, there was acceptable compatibility between the starch and the PVA matrix It has been seen in Scanning Electron Microscopy (SEM), the low presence of voids and agglomerates in this structure has demonstrated that the surface adhesion between the two segments is strong(Rahmah et al.).

Souza et al.explained that tensile strength of starch films decreased by increasing glycerol concentration. By increasing glycerol concentration from (0.17 to 0.75)g/100gm tensile strength value decreased from (6.06 to 2.25)MPa. The presence of glycerol changed the percentage of elongation at break of the film: a diminishing in this property was observed when the glycerol content increased from (0.17 to 0.75) g / 100 g.This is a direct result of the anti plasticizing impact brought about by the high plasticizer content. The utilization of sucrose and invert sugar caused a similar impact ,as they also acted as plasticizers. Veiga-Santos et al.they found that adding sucrose to cassava starch can increase E by up to 2900% contrasted with sucrose free films. As indicated by Teixeira et al. ,this specific behavior can be attributed to the way in which sucrose contains a greater number of OH groups than different sugar, which makes it an increasingly hydrophilic and progressively more effective plasticizing agent.The utilization of sucrose and invert sugar caused a similar impact, as they also acted as plasticizers. Veiga-Santos et al.they found that adding sucrose to cassava starch can expand the E by up to 2900% contrasted with sucrose-free films. As indicated by Teixeira et al. , this particular behavior can be credited to the manner by which sucrose contains a greater number of OH groups than different sugar, which makes it an increasingly hydrophilic and progressively more effective plasticizing agent.

Zakaria et al. explained how the processing temperature influences the mechanical properties of starch-based films. According to the authors, the tensile strength of starch films is inversely proportional to the mixing temperature. By the increasing of temperature from 85°C to 95°C, the tensile strength value diminished from 2.6Mpa to 2.2MPa. The films prepared at a mixing temperature of 85°C showed a tensile strength value of 2.6MPa while the films prepared at 80°C showed a tensile strength value of 2 MPa. This behavior is due to structural changes. During high temperature heating it caused a lower density of the film matrix, thus reducing the tensile strength properties.

Chinma et al revealed how mixing temperature and humidity influence the mechanical properties of cassava starch and soya protein blend film. As inspected by them, humidity and temperature influences the mechanical properties of soy protein and cassava starch film. They arranged the starch film by the casting method. At 50 % relative humidity and temperature they extend somewhere in the range of 10 to 40 °C; The estimations of tensile, elastic modulus and elongation at break of the film are respectively from 20.1 to 24.9 MPa, from 40.8 to 66.7 MPa and from 7.3 to 11.5 % respectively . But with a relative humidity of 80 %, the quality of tensile, the elastic modulus and the elongation at break varied from 19.4 to 23.9 MPa, 39.1 to 65.4 MPa and from 7.1 to 11.2 % respectively.With an increase in temperature to a constant relative humidity, the tensile strength increases slightly marginally yet diminished with an increase in relative humidity .Higher relative humidity and lower temperature caused greater elongation at break. Then again, tensile strength and elastic modulus increased slightly with the increase of temperature at a constant relative humidity ,while the elongation at break has decreased. Decrement of tensile strength and the elastic modulus of edible films with the increment in relative humidity could be ascribed to the increase in the moisture content of the film with the relative humidity. Films those are exposed to higher relative humidity contain more water than films with lower

Mechanical Properties of different starch films				
Starch biocomposite	**Tensile Strength (TS, MPa)**	**Elasticity Modulus (EM, MPa)**	**Strain at Break (ε, %)**	**References**
Corn starch mixed with glycerol and xylitol	2.5–3.6	-	48–63	Fu, Wang, Li, Wei, and Adhikari (2011).
Rice starch mixed with glycerol	1.6–11	21–533	3–60	Dias et al. (2011).
Cassava starch mixed with glycerol	1.4–1.6	5–21	30–101	Muller et al. (2009), Famá, Rojas, Goyanes, Gerschenson (2005).
Corn starch mixed with glycerol and stearic acid	0.2–2.9	3–38	46–91	Pushpadass et al. (2009), Pushpadass et al. (2009).
Potato starch mixed with glycerol	3	45	47	Thunwall, Boldizar, and Rigdahl (2006a); Thunwall, Boldizar, and Rigdahl, M. (2006b).
Rice starch mixed with glycerol	3.2	-	-	Mehyar and Han (2004).
Pea starch mixed with glycerol	4.2	-	-	Mehyar and Han (2004).
Cush-cush yam starch mixed with glycerol	1.88	13.9	19	Gutiérrez et al. (2015).

relative humidity due to the adsorption of moisture .

Antimicrobial properties

Starch has no intrinsic antimicrobial properties, so these properties must be given on it.It can be modified with functional compounds to improve the antimicrobial properties of the edible film. Rungsiri and his co workers used propolis extract (ppl) in Rice Starch/Carboxymethyl Chitosan films to improve the film's antimicrobial properties. According to them, active film contained with 5–10% ppl had an antimicrobial capacity against Gram-positive bacteria (Staphylococcus aureus and Bacillus cereus).This was due to the good compatibility between polymer matrix and ppl. Starch films reinforced with different kinds of antimicrobial essential oil for food packaging industries to extend the shelf life of items and diminish the danger of pathogenic bacteria development on food surfaces (R. Syafiq et al). Amalia et al. suggested that the incorporation of the natural antimicrobial agent on starch/PVA film exhibited antibacterial activity. They used neem (NO) and oregano essential oils (OEO) in Starch/PVA film. They concluded that films containing 6.7% of OEO have good physical properties and good antimicrobial properties. Ana S. Abreu et al.reported that incorporation of closite 30B(C30B) and Ag-Nanoparticles(Ag-NPs) in to starch films caused upgrade of antimicrobial, mechanical and gas barrier properties. The concentration of Ag-NPs was 0.3mM.Addition of natural extracts such as thymol, betalains, anthocyanins, resveratrol and carvacrol, in to starch film showed antimicrobial activity against various bacteria and fungi (Jessica I. Lozano-Navarro et al.). They examined the inclusion of extracts, particularly at concentrations of 2% and 5% v/v, resulted increase of antimicrobial activity. Nafiseh Mohsenabadi et al. demonstrated the inhibitory effects of chitosan (CS)/benzoic acid (BA) nanogel and rosemary essential oil(REO) against S. aureus and the inhibitory effects were increased by encapsulating REO in CS-BA nanogel. Oregano essential oil (OEO) is commonly applied in films based on starch & chitosan to get Satisfactory effects on bactericidal properties. Pelissari et al. applied (OEO) on starch/Chitosan film to improve its antimicrobial properties. They reported that increasing oil concentrations up to 2 % display an inhibitory effect against Gram-positive bacteria and Gram-negative bacteria .

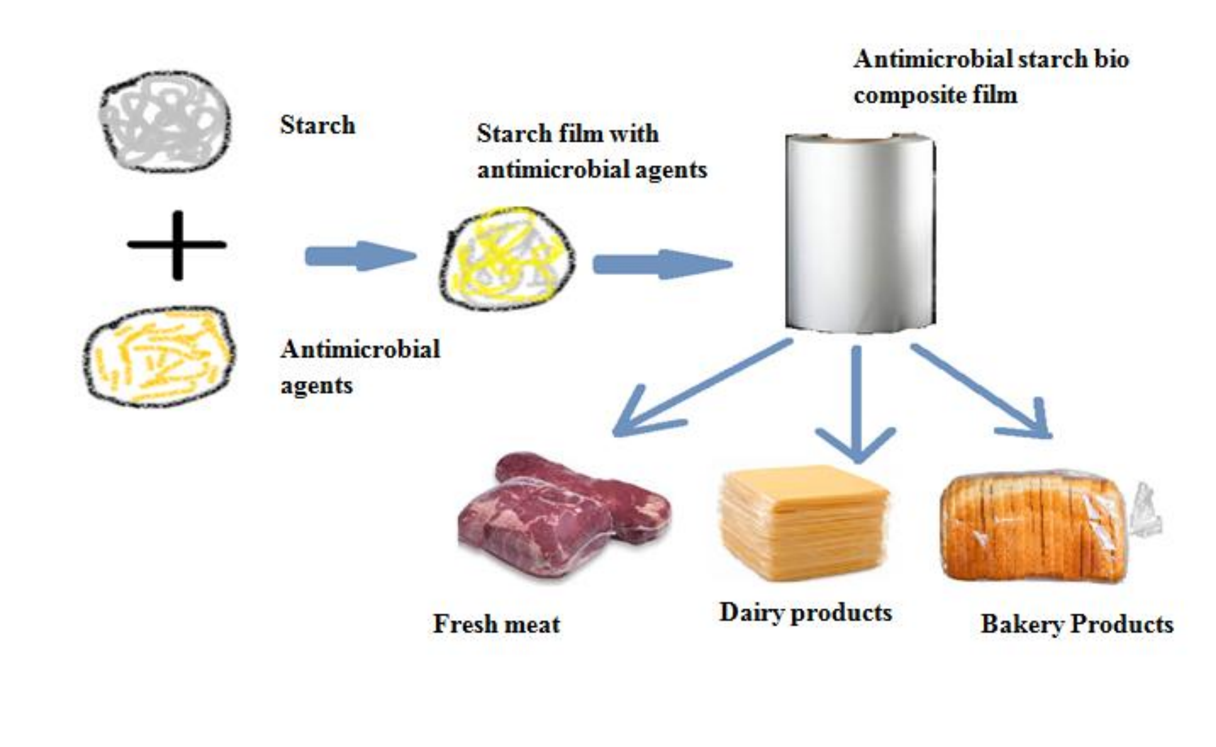

Figure.4.Applications of antimicrobial starch biocomposite film .

Improvement of properties of starch based materials:

Native starches are confined in numerous application since it is insoluble in cold water , high inclination to retrogradation and it can easily undergo syneresis other than the gelling propensity of the pastes . It posses poor processibilty and high hydrophilicity. So as to beat deficiencies of native starches and to achieve demanding technological utilization of starch ;it's modification is necessary.So starch application in food packaging needs various techniques for modification. So to upgrade specific functional properties, numerous functional groups are required to introduce on the backbone of starch. For this starch are modified by various techniques. The techniques are physical, chemical, enzymatical & genetical.

Physical modification of starch

Physical modification of starch broadly utilized in food industry. It is done by alcoholic-alkaline, micronization and drum drying techniques (Majzoobiet al., 2011) . The techniques involve changes of grainy structure of starch in to small crystal shape starch under high pressure, shear &alcoholic-alkaline treatment. Physical modification is carried out by three ways.

a)Pre-gelatinized starches b) Hydrothermal modification c) Heat Moisture Treatment (HMT).

a) Pre-gelatinized starches

Pre-gelatinized starches also called cooked starches that are prepared and dried with the help of a drum. The fundamental properties of PGSs to form modest stable type of suspension that scatter in cold water. PGS can be brought about by three basic types of equipment such as **drum drying, spray drying, and extrusion cooking.** The procedure of drum drying permit in explicit physicochemical modifications of starch granules to increase different surfaces and porous structures which at last impact its functional properties. Drum drying is the most generally used technique at industrial level. Spray drying is a technique generally used in the food industry and these days it is utilized as a microencapsulation strategy for an ingredient. Spray drying treated starch granules having a regular shape with a hollow structure in the middle of the granule (Yan & Zhengbiao, 2010). The dried spherical particles creates by this procedure are generally used to modify the starch physically. Extrusion cooking process involves convert of starch from a granular & semicrystalline material in to a viscous plastic material by the application of pressure, high temperature & shear force.

b)**Hydrothermal modification**

Two hydrothermal treatments, for example, Annealing (ANN) and high moisture treatment (HMT) are utilized to alter the physicochemical properties of starch without losing the respectability of the granule structure. The prominent feature of these strategies is that both physical modifications happen beneath the gelatinization temperature of starch granules or more the glass transition temperature (Tg). Both ANN and HMT need discrete temperature and moisture for the storage of starch during a specific period of time.HMT alludes to the treatment of starch at low moisture level(less 35% w/w) while ANN is utilized to treat starch in abundance water (less 65% w/w) or at intermediate water (40–55% w/w).

c)**Heat-Moisture treatment**

Heat moisture treatment (HMT) known as a hydrothermal technique that comprises of agitation of starch granules over the starch's glass transition temperature(80-140 °C). Numerous tests have revealed that the physical properties of the heat-moisture treated starch rely upon the moisture content, temperature, botanical source, heat source, cooling process and duration of treatment. The large literature on HMT has been analysed by Hoover (2010), Zavareze and Dias (2011), and BeMiller and Huber (2015). There is likewise a general assent that the procedure of HMT at moderately low-moisture content the high temperature achieved changes in both crystalline and amorphous regions of the starch granule because of expanded mobility of starch polysaccharide chain segments. HMT causes changes in starch material, for example, diminishes starch solubility, swelling power, and draining of amylose from the swollen granules. In any case, it expands RVA pasting temperatures, diminishes peak viscosities and breakdown, and expanded hot-paste viscosities. Starch treated with HMT acknowledged an industrial level for the preparation of infant foods.

Chemical modification of starch

Chemical modification is completed by incorporating different functional to the starch molecules, for example, carboxyl, acetyl, and hydroxypropyl that achieves the extraordinarily changed physicochemical properties. The chemical and functional properties of the changed starches rely on the starch source, reaction conditions (reactant concentration, reaction time, pH and the presence of a catalyst), degree of substitution, type and distribution of substituting agent in the starch molecule Chemical modification of the starches is commonly carried out by derivatization such as acetylation, cationization, oxidation , cross linking and acid hydrolysis.

Table shows brief summary of procedure of different chemical modification methods and also properties of modified starch prepared from various techniques of chemical modification methods.

Name of the chemical modification	Methods of modification	Properties	Reference
Cross-linking	Cross-linked starch, prepared by means of incorporation of hydrogen bond with chemical bond in to the starch granule that goes about as a bridge between the starch molecules. There are different cross-connecting reagents, for example, phosphoryl chloride, epichlorohydrin, sodium tripolyphosphate, sodium trimetaphosphate	Decrease in retrogradation rate, Better freeze–thaw stability, increase in gelatinization temperature,better granule stability, Lower swelling power, High heat and shear stabilities	Koo et al.
Grafting	During the grafting process engineered or common polymers grafted in to the backbone of the starch. Grafting is	Film making ,good thermal stability biodegradability , High hydrodynamic volume and	E. Al et al.

	performed by vinyl monomers such as styrene, acrylic acid,acrylamide etc.The grafting mechanisms are free radical, condensation, addition, and ionic mechanisms.	Stronger water absorbency.	
Esterification	The esterification of the starch is carried out by derivatives of acids or carboxylic acids, in the existence of a vast quanity of hydrophilic hydroxyl groups. Generally, there are two sorts of starch esters, inorganic and organic starch esters.	Reduction of sensitivity to humidity , reduction of thermal resistance and biodegradation, better compatibility and reduction of the hydrophilic surface character of the starch film.	R. Santayanon et al.
Etherification	The etherification of the starch is carried out by the reaction between the hydroxyl group in the starch molecule with a reactive substance which includes cationic starch, anionic starch, amphoteric starch and nonionic starch.	Improvement in physio-chemical properties like thermal stability, ion activity, solubility, permeability , strength, higher reaction efficiency and thixotropy	O. S. Lawal et al.
Oxidation	Oxidation of starch is a helpful modification technique to improve starch properties by reacting the starch with a particular amount of oxidant at controlled pH and temperature. Oxidants, for example, sodium hypochlorite, potassium permanganate, peracetic acid, hydrogen peroxide, chromic acid, and nitrogen dioxide have been utilized for the oxidation of starch.	Improve solubility,viscosity, freezing-thawing stability, water binding capacity, light transmittance.	A. O. Oladebeye et al.
Acidification	Modification of starches utilizing mineral acids is a typical kind of modification strategy where the hydroxonium ion attacks the glycosidic oxygen atom and hydrolyzes the glycosidic linkage. It changes the properties of starch without destroying starch granule structure.	Increased solubility,decreased Swelling power,pasting viscosity, gelatinization enthalpy.	N. Atichokudom-chai et al.
Dual	Dual modification of starches incorporates the blend of either chemical and physical modification or chemical and enzymatic modification strategies that make serve to upgrade the pace of derivatization or can improve the level of substitution in certain cases. For the most part, dual modification of starch includes a blend of two chemical modification strategies, for example, cross-linking/acetylation, acetylation/oxidation, or cross-linking/hydroxy propylation.	Combination of properties	Zhou et al.

Enzymatic Modifications

Enzymatic modification of starch has been used as an effective tool to improve their texture, functionality, and nutritional quality of starch by modifying starch in terms of branch chain length , molecular mass, phosphate substitution and disproportionation . Enzymes used in processing of starch are α-amylase, β-amylase, glucoamylase, pullulanase, and isoamylase.

In enzymatic modification method structural changes and processing are mainly focused. An expansion in the starch branch density that implies increase in the extent of long-branch chains and decrement in the extent of short-branch chains caused moderate processing. To modify structure of starch the major reactions used are: (1) hydrolysis of a-1, 4 or a-1, 6 glycosidic linkages, (2) disproportionation by the transfer of glucan moieties, and (3) branching by formation of a-1, 6 glycosidic linkage. The utilizations of enzymatic modification of starch are the elimination of starch retrogradation, the development of the emulsification properties, the favourably branched nanostarch molecule, the prevention of the aging of bakery products.

Genetical modification:

Genetic modification in starch create mutants in genes which control starch structure and quanity.It involve transgenic technology which target enzymes those are involved in starch biosynthesis.Starch modification in terms of amylose-amylopectin ratio is mostly used. Genetic modification is carried out by biotechnology.

1.low amylose content starch:

Inactivation of three starch synthase genes (Granular Bound Starch Synthetase, Starch Synthetase II and Starch Synthetase III), brought about waxy (Wx) locus in plants gives amylose-free phenotype in storage tissues with little amylopectin chains with no clear decrease in absolute starch content. Currently waxy starch from corn or maize is in use but by the introduction of amylase-free potato starch in Europe expands the uses of waxy starch in the market. Waxy potato starches have greatly improved functionality as it gelatinizes effectively, yielding clear pastes with high freeze-thaw stability contrasted with typical potato starch.

2.High amylose content starch:

High-amylose starch in grains is accomplished by altering the expression of different starch branching enzymes, for example, SBEI, SBEIIa, and SBEIIb. In japonica rice and in potato, it was like wise demonstrated that down regulation of expression SBEIIb delivered just a little increment in amylose levels. Anyway it was important to inhibit expression of both SBEI and SBEII to make starches with amylase levels up to just a limit of 27%. (Zhu, Gu, et al., 2012). Further increments in amylose were accomplished by inhibiting the capacity of gene and crossing the double-gene down regulated with higher amylase level (27%).High amylase starch are more resistant to digestion and can be prepared to "resistant starch" which has wholesome advantages.

Formulations of thermoplastic starch

Plasticisers

Native starch based materials processing technology is one of the most important issues in starch processing production due to brittle nature of starch with surface cracks. However, these drawbacks can be resolved by adding an appropriate amount of plasticizer to pure starch to improve its workability. The character of plasticizers is to build the flexibility and workability of the starch film by diminishing the intramolecular hydrogen bonding between starch particles. As an outcome, molecular mobility expands, crystallinity diminishes and glass transition temperature additionally declines. The most usually utilized plasticizers for starch-based films plasticizers are hydrophilic low molecular weight compounds, for example, glycerol and sorbitol. A few works have been distributed about the impact of various plasticizers on films properties dependent on different starch origins.

List of different Plasticizers used in biodegradable film from biomass products.

Type of film	***System of application***	***Plasticizer***	***Refs.***
Polysaccharide based film	Potato starch	Glycerol	Funke U et al.
	Citric acid-modified pea starch (CAPS) and citric acid-modified rice starch (CARS)	Glycerol and ethylene glycol	Ma X et al.
	Corn starch	Glycerol, formamide, , acetamide anhydrous glucose and urea Sorbitol and Glycerol . Caproic acid, lauric acid and glycerol triacetate (triacetin). Glycerol and amino acids.	Fringant C et al. Ma X & Yu J Garcia MA et al.
	Oat starch	Sorbitol, glycerol and urea, sucrose and glycerol-sorbitol mixture	Galdeano MC et al.
	Cassava starch	Glycerol	Bergo PVA et al.
	cellulose acetates	Residual xylan acetate	Shaikh HM et al.
	ϒ-Carrageenan edible films	Glycerol and water	Karbowiak et al.
Protein based film	Whey protein	Glycerol and sorbitol	Kim SJ & Ustunol Z
	Whey gluten	Glycerin	Sobral PJA et al.
	Fish skin protein	Fatty acids (FA) and sucrose esters (FASE)	Jongjareonrak A et al.

	Caseinate-pullulan	Water and sorbitol	Kristo E & Biliaderis CG
	Pigskin gelatin	Glycerol	Bergo P & Sobral PJA
	Sunflower protein	Saturated fatty acids (FA)	Orliac O et al.
	Bovine gelatin	Fatty acids	Bertan LC et al.
Film from microbial sources	Poly(3-hydroxybutyrate) (PHB)	lauric acid, Dodecanol, tributyrin and trilaurin	Yoshie N et al.
	Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV)	epoxidized soybean oil (ESO), Soybean oil (SO), dibutyl phthalate (DBP) and triethyl citrate (TEC)	Choi JS & Park WH

Fillers(Including Nanofillers)

The role of fillers is to modify the properties of the starch film. Cellulose fibers, Micro- or nanosized starch, montmorillonite, carbon nanotubes, metal chalcogenides and metal oxides are some fillers examples which are incorporated into the starch matrices. One of the important parameter in case of nanofiller is the filler particle size. Incorporation of nanofillers in starch matrix caused various improvement in properties such as electrical, mechanical, thermal, antimicrobial and flame resistant.

List of nanofillers that have been used for food packaging applications.

Classification	Type of Nanofillers	Properties enhanced	References
Clay nanofillers	MMT, Hal etc.	Good mechanical stability Thermal stability	Fathi Achachlouei & B.; Zahedi
Organic Nanofillers	Nanocellulose	Antibacterial effect Thermal stability Blood compatibility Good mechanical stability	Poonguzhali et al.
	Chitosan nanoparticles	Bio degradability Low toxicity Antimicrobial activity	Yu, S et al.
Inorganic Nanofillers	SeNPs	Antioxidant activity	Kalishwaralal et al.
	TiO2 NPs	Antimicrobial activity Photocatalytic activity	Alizadeh-Sani et al.
	ZnO NPs	Antifungal effect Antimicrobial effect Thermal conductivity	Noshirvani et al.
	SNPs	Antimicrobial effect	Shankar, S et al.
Carbon Nanofillers	Graphene, graphene oxide etc	Lightweight Processing benefits, resistance to corrosion Extraordinary electrical, mechanical, and thermal properties .	Wang, C et al.

Future of starch film in food packaging applications:

TPS has great potential to use in large scale production for biodegradable packaging materials. It has control to establish an inviolable role between the assembling of plastics and different bioproducts to a great extent since it is rich, inexhaustible, and economical. In the future It can be utilized with respect to the wrapping of food, isolating layers between segments of food, heat-fixed to frame sacks, pockets, sachets, or packs to contain dry foods or preweighed ingredients. (**Zhang et al.).**But starch-based films have some strong limitations such as they are sensitivity to moisture and brittle. So as to beat these limitations, starch has been mixed with various plasticizers, additives and nanoparticles. But according to technological point of view to increase properties of starch based film there is more research is required for new formulation of thermoplastic starch method and also a good understanding of processing of thermoplastic starch. Many work has been done on starch blended with biodegradable and non-biodegradable polymer. But very less interest was shown on starch blended with engineering polymers(PC,ABS,PET,PBT,PA).So it is a good future platform where material with high performance with biodegradability can be made.

V. CONCLUSION:

Until this point, the outcomes got in polymer science have expanded our insight. Starch as a packaging material is socially mindful, monetarily suitable and is proposed as a significant device to conquer the current difficulties related to packaging material and, subsequently, therefore expanded toughness, quality, security, and nourishment assurance. A few modification techniques have been created to deliver films/coatings with improved preparing abilities and have increased more prominent enthusiasm for modern and scholastic research. The starch film indicated great development properties, for example, air, moisture barrier, thermal sealing capacity, and so on. The addition of additives in starch films is important to deliver an increasingly ductile and flexible material, improving the treatment of the film. Methodologies to improve the properties of starch-based plastics, for example, blending starch with different polymers, utilizing starch in composite materials and utilizing starch as fermentation. The raw material for the production of different biopolymers has been effective in creating legitimate substitutes for petroleum-based plastics. Starch prospects in the packaging business keep on being more brilliant. The economical plastics market advances advancement and improvement.

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