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*Corresponding author: Adil Gani,
Department of Food Science and
Technology, University of Kashmir,
Srinagar 190006, Jammu and Kashmir,
India
E-mail: adil.gani@gmail.com

Reviewing editor:
Fatih Yildiz, Middle East Technical
University, Turkey

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FOOD SCIENCE & TECHNOLOGY | REVIEW ARTICLE

A review of the recent advances in starch as active and nanocomposite packaging films

Umar Shah¹, Adil Gani^{1*}, Bilal Ahmad Ashwar¹, Asima Shah¹, Mudasir Ahmad¹, Asir Gani¹, Idrees A. Wani¹ and F.A. Masoodi¹

Abstract: Technological advances have led to increased constraints regarding food packaging due to environmental issues, consumer health concerns, and economic restrictions associated therewith. Hence, food scientists and technologists are now more focused on developing biopolymer packages. Starch satisfies all the principle aspects, making it a promising raw material for edible coatings/films. Starch as a package material has grabbed much attention both at academic as well as industrial levels. Besides this, the role of various plasticizers, polys, sugars, and wetting agents are discussed and their importance in packaging industries. Herein, the role of starch as packaging material and nanofillers/composites is discussed in detail. The review summons a comprehensive and current overview of the widely available information and recent advances in starch film packaging.

Subjects: Bioscience; Environmental Studies & Management; Food Science & Technology

Keywords: biopolymer; biodegradable; nanofillers; bio-nanocomposite; food packaging

1. Introduction

Starch is unique among the carbohydrates because of the discrete particles called granules. Starch granules are insoluble; they hydrate only slightly in cold water. As a result, they can be dispersed in water producing low-viscosity slurries that can be easily mixed and pumped, even at concentrations greater than 35%. The viscosity-building power or thickening power of starch is observed only when the slurry of granules is cooled. Among the renewable sources with film-forming capability, starch

ABOUT THE AUTHORS

Umar Shah completed his MTech in Food Technology from Amity University U.P. Currently, he is a research associate at the Department of Food Science and Technology, University of Kashmir. He has attended various international and national conferences besides having various research papers in Elsevier, Springer, Taylor and Francis, Wiley, and other peer journals.

Adil Gani is an assistant professor at the Department of Food Science and Technology, University of Kashmir. He has attended various international and national conferences and has more than 40 publications in his credit.

F.A. Masoodi is professor and head at the Department of Food Science and Technology. He has attended various international and national conferences and has more than 90 publications in his credit.

PUBLIC INTEREST STATEMENT

Food packaging is one of the fastest emerging technologies. Starch films have grabbed more and more attention/attraction both at industrial and academic levels. This review summons a comprehensive and current overview of the widely available information and recent advances in starch film packaging.

satisfies all the principle aspects such as easy availability, high extraction yield, biodegradability, and biocompatibility, making it a promising product for edible coatings/films (Babak, Hadi, & Entezami, 2011; Falguera, Quintero, Jiménez, Muñoz, & Ibarz, 2011; Kowalczyk & Baraniak, 2014; Souza, Benze, Ferrão, Ditchfield, & Coelho, 2012). Starch films are odorless, tasteless, colorless, nontoxic, biologically absorbable, semi-permeable to carbon dioxide, moisture, oxygen, lipids, and flavor components. The properties of starch film are similar to the effect that is promoted by storage under controlled or modified atmosphere and can be attributed to its chemical composition. Starch granules are composed of a mixture of two polymers—amylose and amylopectin. These polymers have the same basic structure but differ in their length and degree of branching, which ultimately affect the physiochemical properties. Amylose is essentially a linear polysaccharide or sparsely branched with α (1–4) bonds with a molecular weight of 10^5 – 10^6 and can have a degree of polymerization (DP) as high as 600. Amylopectin is a highly branched polymer with a molecular weight of 10^7 – 10^9 and α (1–4) (around 95%) and α (1–6) (around 5%) linkage and with a pending chain of DP~15, which is responsible for materials' crystallinity and this structure affects the physical and biological properties (Ashwar, Gani, Shah, Wani, & Masoodi, 2015; Pérez, Baldwin, & Gallant, 2009). Starch is one among the most important functional polymers. The functional role of starch in food products is enormous including being used as an adhesive, for binding, clouding, dusting, film forming, foam strengthening, gelling, glazing, moisture retaining, stabilizing, texturizing, and thickening applications (Damodaran, Parkin, & Fennema, 2008, *Fennema's Food Chemistry*). Starch is not a real thermoplastic polymer but by addition of water can be processed after gelatinization (González, Retegi, González, Eceiza, & Gabilondo, 2015). Water improves the conductivity by improving the movement of starch chains. The three-dimensional architecture and semi-crystalline nature of starch are disrupted by heating in water consequently causing phase transition from ordered granular structure into disordered state in water. The high water addition causes crystallites in starch and might be pulled apart by swelling and low water content causes high steric hindrance (Xie, Pollet, & Halley, 2013). The review summons the up-to-date information regarding recent advances in starch as active and nanocomposite packaging films.

1.1. Starch as a packaging material

Foods are packed in different packaging materials, most belonging to the following classes: metals, glass, paper, and polymers. Some of the packaging materials consist of a combination of two or more materials of these classes. A good food package involves four important functions of protection, communication, containment, and convenience to maintain the safety and quality of food products during storage and transportation and also to extend products' shelf life by preventing unfavorable conditions such as spoilage, micro-organisms, chemical contamination, light, moisture, oxygen, and external force. Hence, the food package should prevent moisture gain or loss, microbial contamination, and should act as a barrier against permeation of oxygen, carbon dioxide, water vapor, and other volatile compounds such as flavors and taints besides their basic properties like mechanical, optical, and thermal properties. Due to the versatility and low cost, polymers are the most important packaging materials for foods. Their permeability and oxygen are important criteria for selection of polymer films as packaging materials (Berk, 2013). The most popular packages are made of plastic polymers, e.g. low-density polyethylene, high-density polyethylene, and polyvinylchloride. However, the treatment of these waste plastics is a serious problem because they are derived from petroleum products and cause problems during waste disposal (Avella et al., 2005). Therefore, biodegradable packaging is done to replace the synthetic and non-biodegradable packaging to meet the increasing demand for sustainability and environmental safety in all food applications. Natural biopolymer films are derived from renewable raw materials, and are developed into eco-friendly food packaging materials due to their biodegradability. The natural materials such as polysaccharides, lipids, and proteins and their derivatives which are abundantly available are generally used for their preparation. The natural raw material, polysaccharides, is the earliest and most extensively studied material for bio-packaging.

2. Edible coatings

Consumers demand higher quality and longer shelf life foods, while reducing disposable packaging materials and increasing recyclability. Such demands have caused increased interest in edible and bio-degradable films or materials which serve as mass transfer barriers to moisture, oxygen, carbon dioxide, lipids, flavor, and aroma between the food component and surrounding atmosphere and are used to extend shelf life and improve the quality of almost any food system. Such films have the ability of decreasing the amounts of non-renewable conventional synthetic polymer packaging materials and use ingredients of agriculturally derived products. Edible packages are edible coatings and films which are used because of their special ability of increasing the shelf life of many products (Nunes et al., 2013; Wu, Zhong, Li, Shoemaker, & Xia, 2013). Edible film is defined as a thin layer of material, which can be eaten as a part of whole product, providing a barrier to mass transfer (moisture, gas, flavors, etc.) between food and environment or in the food itself (Broumand, Emam-Djomeh, Hamed, & Razavi, 2011). Among the biopolymers, the bio-nanocomposite material for food packaging applications mostly is starch and its derivatives (Tang, Alavi, & Herald, 2008). They are edible and are safe as any food packaging material. However, starch has low mechanical properties which can be improved with additives such as plasticizers and nanofillers (Sorrentino, Gorrasi, & Vittoria, 2007). However, some of the drawbacks of biopolymers as food packaging materials compared to the non-bio-degradable materials include poor mechanical properties (e.g. low tensile strength), high hygroscopic capacity, rigidity, brittle character, and barrier properties (e.g. high water permeability) (Othman, 2014). In particular, brittleness, low heat distortion temperature, high gas and vapor permeability, and poor resistance to long processing operations have strongly limited their applications (Rhim, Park, & Ha, 2013). To overcome these limitations, starch is blended with other polysaccharides and plasticizers which resulted in good film-forming ability. It is a well-known fact that synthetic or bio-degradable packaging films have better mechanical properties, including tensile strength and elongation, but the blending of starch with other polysaccharides, plasticizers, and additives have met the demanding mechanical properties of packaging.

2.1. Plasticizer

Addition of water (universal solvent) as a plasticizer has poor mechanical properties, reduces viscosity during thermal processing, and varies with humidity. It causes brittleness in film at lower humidity and softens films at high humidity. The addition of only one plasticizer reduces the film flexibility by internal hydrogen bonding between polymer chains, thereby increasing intermolecular space. For suitable gelatinization conditions, various plasticizers have been used in combination with water. The use of glycerin as a plasticizer has an important characteristic, that is, the missing of retrogradation, which normally occurs with glycerol and other plasticizers (Battezzato, Bocchini, Nicola, Martini, & Frache, 2015). Glycerin is used mainly to maintain moisture level for a continuous film casting. Glycerol, as a plasticizer has been widely used in edible films and is becoming an attractive plasticizer because it is a bio-diesel residue. Among the various steps of edible film preparation, drying of the film is important because the drying method and conditions affect the properties of bio-film. Also, the addition of glycerol shows a decrease in moisture diffusion values during drying of plasticized edible films. While thermally processing of native starch, addition of plasticizer is important as the decomposition temperature is lower than its melting temperature before gelatinization. Addition of universal solvent (water) as a plasticizer has poor mechanical properties and varies with humidity (brittleness at lower humidity and soft at high humidity) but reduces viscosity during thermal processing. The effect of water as a plasticizer depends on various parameters such as processing history and the presence of other additives. In order to approach the conditions for suitable gelatinization, various plasticizers have been used in combination with water.

2.2. Polyols and sugars

Polyols (polyhydric alcohols which include propylene glycol, glycerin, and sorbitol) as additives may be added in the formulation to modify mechanical properties and provide significant changes in flexibility, avoid pores, and extensibility of the polymeric matrix. With addition of plasticizer, flexibility of a film can be reduced by internal hydrogen bonding between polymer chains, thereby increasing intermolecular space (Liu, Xie, Yu, & Chen, 2009). The crystalline nature of the film depends on the grade of

amylose dissolution and amount of hydrogen bonds formed within the film. One of the important properties of the film is solubility in water which is useful for encapsulation of food or additives and for film formation. Use of glycerin as a plasticizer in starch-based films has been generally recognized as safe (GRAS) status by FDA because of the non-sweet and inexpensive nature. The most important characteristic of this plasticizer is the missing of retrogradation, fact that normally occurs with glycerol and other plasticizer agents (Battezzore et al., 2015). The films without plasticizers are relatively brittle and seal separated instantly after force is applied without rupture. Glycerin is used as a plasticizer or humectants to maintain an adequate moisture level for a continuous film casting. Keeping the film hydrated assures adequate flexibility and resiliency. Cracks are developed in starch-based edible films when subjected to relative humidity below 20–25% and use of glycerin and other polyols can lower tolerances to 10–15% relative humidity. Mechanical properties and transparency of the edible films are improved by the application of glycerol. Starch-based films with glycerol have lower polymer chains. The plasticizers (glycerol) have a function: to improve flexibility and cause changes in the structure network of starch, as matrix becomes less dense and under tension facilitates movement of polymer chains. Glycerol and urea could form more stable and stronger hydrogen bonds with water and starch than a single plasticizer such as glycerol (Rahman, Lee, Rahmat, & Samad, 2000; Sun, Wang, Kadouh, & Zhou, 2010). Sucrose had demonstrated a higher plasticizing efficacy when comparing to sorbitol and glycerol. The addition of sorbates to edible films results in minimizing surface microbial contamination as they are active against yeast, molds, and many bacteria and are considered as GRAS additives (Barzegar, Azizi, Barzegar, & Hamidi-Esfahani, 2014). Broadly speaking, starch-based films are very strong and the strength is directly dependent upon the moisture content of the film and relative humidity of the environment in which they are stored. Due to the negative effect of plasticizers on the barrier and mechanical properties of films, the amount of plasticizer needs to be optimized (de La Cruz Garcia, Lopez Hernandez, & Simal Lozano, 2000). The concentration of the plasticizers used in starch-based formulations is described by various scientists, e.g. glycerol and sorbitol are used with a typical concentration ranging between 0 and 50 g/L. The higher values of the WVP in coatings without plasticizer in comparison to films with plasticizer as in unplasticized coating lead to pores and cracks. The magnitude of WVP in protein and other polysaccharide films is higher in comparison to starch films (Parris, Dickey, Kurantz, & Craig, 2005). Low molecular weight materials such as sugars are prone to act as effective plasticizers in starch-based films but make films more brittle and glass like. The addition of sugars to starch-based matrices considerably changes hydrogen bond network and consequently modulates matrix mobility. The similar dynamic effects were seen in case of sugar as additives while comparing it with other additives (gelatin, amylose, or xanthan gum) and concluded that polymers such as amylose or xanthan gum enhance local mobility of sugars, while small polyols such as glycerol, maltitol, and lactitol suppress local mobility in sugars. Use of plasticizers in starch modification decreases intermolecular attraction, increases polymer mobility, increases elongation, and decreases tensile strength as glycerol content increases. The starch-based coating with plasticizer is homogeneous and covers the whole surface of the fruit, and coatings without plasticizers are brittle and possess undesirable cracks. Oxidized starch films with glycerol are usually regarded as hydrophilic material (Kuakpetoon & Wang, 2006). The oxidized starch produces low viscous and excellent film-forming properties. The mobility of the chain during crystallization process and ability of the amylose chain determine the degree of crystallinity in the starch films. Slow drying of films causes greater crystalline fraction and faster water evaporation can increase amorphous phase and the presence of the plasticizers (sorbate) limits crystal growth and recrystallization of starch (Fama, Rojas, Ana, & Gerschenson, 2005). Isosorbide as a green plasticizer has an important characteristic of missing retrogradation, fact that normally occurs with glycerol and other plasticizer agents (Battezzore et al., 2015).

2.3. Lipids and waxes

Addition of lipids causes improvement in barrier, mechanical, and optical properties of the film due to the change in inner structure and film surface (Jiménez, Fabra, Talens, & Chiralt, 2013; Jost, Kobsik, Schmid, & Noller, 2014). Addition of the waxes improves moisture barrier properties of the film. Coating of lipid nanolayer (sunflower oil) with hydrophilic film (starch) showed an increase in tensile strength and also low water diffusion coefficients since the structure and the hydrophobic nature of the lipids and oils restrict the migration of gas and vapors (Slavutsky & Bertuzzi, 2015).

2.4. Wetting agents and emulsifiers

Lipids commonly used for films and coatings are stearic acid, palmitic acid, soybean oil, sunflower oil, etc. as they have lower vapor permeability than natural waxes. On addition of lipids as additive to starch films, the inner structure as well as film surface gets affected and consequently causes improvement in barrier, mechanical, and optical properties of the resulting film. Slavutsky and Bertuzzi (2015) reported improvement in water barrier properties while incorporating lipid nanolayer in film, as permeation phenomenon is controlled by diffusion through the hydrophobic nanolayer. The addition of high melting point waxes has prone to be effective in improving moisture barrier properties of the resulting film and by adding 20% edible oil to starch-based coating solution generally extends shelf life of the coating layer in high oil foods since the structure and the hydrophobic nature of the lipids and oils restrict migration of the gas and vapors (Slavutsky & Bertuzzi, 2015). Lipid nanolayer (sunflower oil) was coated with hydrophilic film (starch) and the laminated film showed increase in tensile strength, low water diffusion coefficients, Diffusivity decrease with water activity and Young module with a decrease in elongation in relation to starch-based films (Slavutsky & Bertuzzi, 2015). The hydrophilic films are moisture sensitivity and can be reduced by adding hydrophobic material (de La Cruz Garcia et al., 2000). The use of long-chain saturated fatty acids and fatty alcohols reduces the optic properties of the films. Coating transparency and vapor permeability are affected by the addition of lipids but have a limitation as it offers after taste. Drawbacks of lipid emulsion-based films are that they have low lipid melting temperatures, solvent volatilization from the structural network, and strong effect of emulsion droplet size and distribution, polarity, degree of saturation, and polymorphism of lipid components on water barrier properties and mechanical properties of films and coatings.

2.5. Antioxidants

Organic acids, phenolic acids, terpenes, tocopherols, carotenoids, vitamins, etc. are natural antioxidants and have been used in starch films to improve product stability to prolong storage period (Ashwar et al., 2014; Inam u Nisa et al., 2015). Addition of citric acid in starch films produces a denser structure with a reduction in waste loss and permeability by undergoing cross-linking reactions. Addition of extract of *Nymphaea Mexicana* as an antioxidant (Shah et al., 2014) can enhance the properties of the film. Buckwheat is high in phenolic content (Jan et al., 2015) and can be a possible candidate in film making with antioxidant properties. Ahmad et al. (2014) suggested that green tea has high polyphenolic content and can be used in film making.

3. Biopolymers and bio-nanocomposite

The polymers consist of monomeric units that are covalently bonded, forming chain-like molecules. The prefix “bio” denotes that biopolymers are bio-degradable. Biopolymers have the capability to be degraded through the action of naturally occurring organisms leaving behind organic byproducts as CO₂ and H₂O with no toxic or no environmentally harmful residue (Liu et al., 2014). Biopolymers can be categorized into four groups depending on the origin of biopolymers, which include:

- (1) Natural biopolymers, extracted from biomass (e.g. agro resources),
- (2) Synthetic biopolymers, from biomass microbial production or fermentation (e.g. polyhydroxy alkanates (PHA)),
- (3) Synthetic biopolymers, conventionally and chemically synthesized from biomass (e.g. Poly lactic acid (PLA)), and
- (4) Synthetic biopolymers, conventionally and chemically synthesized from petroleum products.

The first three groups are derived from renewable resources, while the last group is derived from petroleum.

Nanocomposite was established to improve the mechanical, thermal, and barrier properties of biopolymers and to extend the application field. Bio-nanocomposites consist of a biopolymer matrix incorporated with nanoparticles (fillers) having at least one dimension in 1–100 nm range. They are a new class of materials having improved properties as compared to the biopolymers due to the high

surface area and aspect ratio of nanoparticles. Therefore, bio-nanocomposites for food packaging films have been developed with improved mechanical, barrier, rheological, and thermal properties (Rhim et al., 2013). The nano-sized fillers or the nanoparticles play an important structural role while acting as a reinforcement to improve the mechanical and barrier properties of the matrix. The matrix tension is transferred to nanofillers through the boundary between them (Arfat, Benjakul, Prodpran, Sumpavapol, & Songtipya, 2014; Kanmani & Rhim, 2014). The incorporation of nanoparticles such as silicate, clay, and titanium dioxide (TiO₂) to biopolymers not only improves their mechanical and barrier properties but also adds antimicrobial, biosensor, and oxygen-scavenging properties (Rhim et al., 2013). The nanocomposite acts as both an active food package and smart food package. It acts as an active food package by interacting with food and releasing antimicrobial agent, antioxidant agents as beneficial compounds, and oxygen or water vapor as unfavorable elements. And in smart packaging, it shows the property of packaged food such as microbial contamination or expiry date and uses some mechanism to register and convey information about the quality or safety of food. Among the different nanoparticles, TiO₂ nanoparticles are widely used because they are cheap, non-toxic, and photostable with recommended safe dosage. Metal oxides play an increasingly important role in the development of modern technologies. One such material is Titanium dioxide (TiO₂). When nano-TiO₂ particles are incorporated into a polymer matrix such as packaging material, they will provide protection against micro-organisms as well as odor, staining deterioration, and allergens due to the presence of radiation of relatively low wavelength near UV region. When used in food packaging, nano-TiO₂ could be able to withstand the stress of thermal food processing, transportation, and storage. This material exhibits high transparency, very good thermal, chemical, and mechanical activities, and high photocatalytic activity. TiO₂ is also one of the basic “high index” material applied in the construction of optical coatings. These coatings are required to fulfill other functions, e.g. protective. But this improves requirements regarding their mechanical properties, especially hardness, abrasion/scratch resistance, and flexibility (Mazur, Morgiel, Wojcieszak, Kaczmarek, & Kalisz, 2015). The hardness of the film is usually many times smaller than that of bulk ones. However, it was found that hardness can be increased by depositing thin films with a dense structure, while for nanomaterials, it is possible to achieve hardness even greater as compared to bulk ones. This is because the hardness of material increases when the grain size decreases, with the maximum in the nanometer range (Hall-Petch-effect) (Gao et al., 2003).

3.1. Nanoparticles or nanofillers

It has been suggested that the inherent limitations of biopolymer-based packaging materials can be overcome by nanocomposite technology. Nanocomposites exhibit increased barrier properties, increased mechanical strength, and improved heat resistance compared to their native polymers and conventional composites (Rhim et al., 2013). At nanoscale level, the size of nanoparticles, or filler is significantly reduced, leading to the dramatic increase in the surface area of fillers. This is desired because bio-nanocomposites depend on high surface area of the nanoparticles which results in a large interfacial area or boundary area between the biopolymers and nanoparticles. The large interface enabled the modification of molecular mobility, the relaxation behavior besides the mechanical, thermal, and barrier properties of bio-nanocomposites. The bio-nanocomposite materials are especially designed to have the ability to endure the mechanical and thermal stress of food packages during food processing, transportation, and storage (Rhim et al., 2013). Many types of nano-sized particles (<100 nm) have been used to enhance the performance of biopolymers. The nano-sized fillers can be either organic or inorganic such as clay (e.g. montmorillonite (MMT), natural biopolymers (chitosan), natural antimicrobial agents (nisin), metal (silver), and metal oxides (TiO₂). Usually, low amount of fillers or particles (<5%) is adequate for an improvement in the biopolymer properties (Rhim et al., 2013). The mechanical properties are generally dependent on the amount of nanofillers. Many studies have shown that the tensile strength and modulus of bio-nanocomposite materials increase, while elongation at break decreases with an increase in nanofiller amount. The increase in mechanical properties of bio-nanocomposite materials is due to the high rigidity of nanofillers as well as excellent affinity between biopolymer and nanofiller at the interface (Rhim et al., 2013). The incorporation of rigid nanofillers at the interfacial interaction forms the rigid bio-nanocomposite materials, thus enhancing the thermal properties of bio-nanocomposite materials. Studies have shown that improvement in

barrier properties was dependent on types and amount of nanoparticles used as well as the aspect ratio of nanoparticles (Rhim et al., 2013). Between these two, aspect ratio was found to have a major effect on barrier properties of nanocomposite materials. Due to their excellent barrier properties, nanocomposite materials in food packaging applications could lead to considerable shelf life enhancement of food product. The nanoparticle incorporation not only enhances the mechanical, thermal, and barrier properties of biopolymers but offers other functions and applications in food packaging such as antimicrobial agent, biosensors, and oxygen scavenger. The nanoparticle incorporation exhibiting the antimicrobial properties could enhance food safety by controlling the growth and invasion as well as killing bacteria and pathogenic micro-organisms in food. The large surface area of nanoparticles permits more micro-organisms to attach to the nanoparticles and thus increase the microbial efficiency of nanocomposite materials. Some of the nanoparticles exhibit oxygen-scavenging properties. The incorporation of O₂ scavenger nanoparticle helps reduce and maintain O₂ level. This is important because high concentration of O₂ can lead to deterioration of some foods. Packaging the food products using materials with O₂-scavenging properties can limit oxidation and help keep the freshness of food. TiO₂ is one of the nanoparticles that exhibits O₂ scavenger properties under UV light.

4. Conclusion

To date, achievements in polymer science have increased our knowledge. Starch as a packaging material is socially responsible, economically viable, and is suggested as an important tool to overcome existing challenges that are associated with packaging material and consequently resulting in enhanced shelf life, quality, safety, and security of foods. Various modification methods have been developed to produce films/coatings with improved forming capacities and have gained increased interests in both industrial and academic research. The starch film showed excellent forming properties like air, moisture barrier, heat-sealing capacity, etc. The addition of additives in starch films is required to produce a more ductile and flexible material, improving film handling. Starch consisting of crystalline and amorphous domains is a possible candidate to acts an organic nanofiller because the amorphous domains can be removed under certain conditions.

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Competing interests

The authors declare no competing interest.

Author details

Umar Shah¹

E-mail: umarsopore.88@gmail.com

Adil Gani¹

E-mail: adil.gani@gmail.com

Bilal Ahmad Ashwar¹

E-mail: ashwar20@gmail.com

Asima Shah¹

E-mail: shahasimaau2@gmail.com

Mudasir Ahmad¹

E-mail: mudasirahmad63@yahoo.in

Asir Gani¹

Idrees A. Wani¹

E-mail: idwani07@gmail.com

F.A. Masoodi¹

E-mail: masoodi_fa@yahoo.co.in

¹ Department of Food Science and Technology, University of Kashmir, Srinagar 190006, Jammu and Kashmir, India.

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