

Review Article

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A Review on Biodegradable Films

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ABSTRACT

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In Today's scenario, various food industries use harmful packaging materials like plastics, metal, paper and combination of more than one material or composites. Approximately, 60% of plastics are being used in packaging and almost half of that is used to pack food-products, which is very dangerous for human health. Plastic is non-biodegradable and is not safe for food packaging and causes various health hazards. Plastic also reduces moisture and oxygen transfer rate of soil and deteriorates the quality of soil. To get over this problem, there is an urgent need to develop packaging films, which is biodegradable and safe for food packaging. Starch based edible films are biodegradable and also increase the shelf life of food products if incorporated with essential oils.

Introduction

Food packaging is concerned with the preservation and protection of foods and their raw materials, particularly from oxidative and microbial spoilage and also extends their shelf-life. Biodegradable films degrade naturally. Among all the natural polymers, starch has been considered as one of the most promising candidate for future material, because of its attractive combination of price, abundance and renewable in addition to biodegradability. All the plant seeds and tubers contain starch, which is predominantly

present as amylose and amylopectin. Foods that are high in starch include bread, grains, cereals, rice, potato, peas, corn etc. Starch exhibits thermoplastic behaviour (Choi *et al.*, 2007).

The essential oils like cinnamaldehyde, lemongrass oil, clove oil, peppermint oil etc are traditionally being used in food and medicines due to their antimicrobial effects. Cinnamaldehyde is also used as fungicide (Aliabadi *et al.*, 2017). Clove oil is effective against *Escherichia coli*, *Salmonella typhimurium*, *Staphylococcus aureus* and

Listeria monocytogens (Bharath *et al.*, 2017). Lemongrass oil is used as aromatherapy to relieve muscle pain, externally to kill bacteria etc. Lemongrass oil is effective against *Aeromonas veronii*, *Enterococcus faecalis*, *Salmonella enteri* etc. Gram positive organisms are more sensitive to lemongrass oil as compared to Gram negative bacteria (Naik *et al.*, 2010). Antimicrobial activity of peppermint oil against some Gram positive and Gram negative bacteria is explained by Singh *et al.*, 2015.

The biodegradable packaging films with antimicrobial properties can be developed with the addition of these essential oils. The developed antimicrobial films can be used to enhance the storage stability of different perishable foods.

Biodegradable films

Researchers, drug and food industries have shown great interest in the development of biodegradable films since 1980s due to the fact that it can substitute traditional plastic films. These biodegradable films not only enhance the quality of food but also act as barrier for gas, moisture and provide protection to food product after primary package is opened (Kim and Ustunol, 2001).

Various sources can be used in the production of biodegradable films like polysaccharides, proteins, lipids or combination of these. Among these, protein based biodegradable films are found to be attractive with better mechanical and gas barrier properties as compared to lipids and polysaccharides (Ou *et al.*, 2004).

Polysaccharide films

Polysaccharides such as cellulose, chitosan, starch, pectin and alginate are used to form films with good barrier properties against

gases such as oxygen and carbon dioxide. Starch is one of the promising raw materials for the production of biodegradable plastics because of its low cost, its availability as a renewable resource and also its degradation products are innocuous (Mollah *et al.*, 2016). The tensile strength values of starch/chitosan based films were comparable to high-density polyethylene films (Cazon *et al.*, 2017). Chitosan has also been extensively used in films due to its ability to inhibit the bacterial and fungal pathogens growth as it interferes with the negatively charged residues of macromolecules exposed on the fungal cell surface and changes the permeability of the plasma membrane (Romanazzi *et al.*, 2002). Addition of fatty acids was found to be effective in enhancing the antimicrobial properties of chitosan (Dos santos *et al.*, 2012).

Dias *et al.*, (2010) formulated biodegradable films from rice starch, rice flour and characterized their physicochemical, microscopic and mechanical properties. Films from rice starch and rice flour were prepared by casting with glycerol or sorbitol as plasticizer. Scanning electron microscope analysis of starch and flour films revealed compact structures and had comparable mechanical properties. However, water vapour permeabilities were two times higher for rice flour films than those of starch based films. Films with sorbitol were less permeable to water and more rigid while films with glycerol are more plasticized and have poorer water vapour barrier properties. Mollah *et al.*, (2016) formulated biodegradable starch-based chitosan reinforced composite polymeric films by casting. The chitosan content in the films varied from 20% to 80% (w/w).

Tensile strength was improved significantly with the addition of chitosan but elongation at break of the composites decreased. Tensile strength of the composites raised more with

the addition of the *Acacia catechu* content in the films that varied from 0.05% to 0.2% (w/w). The better thermal stability of this prepared film was confirmed by thermogravimetric analysis.

Surface morphologies of the composites were examined by scanning electron microscope suggested sufficient homogenization of starch, chitosan and *Acacia catechu*. Water uptake was found lower for final composites in comparison to starch/chitosan and chitosan films. The developed films intended to use as the alternative of synthetic non-biodegradable coloured packaging films.

Lipid films

The efficiency of lipid materials in film formation depends on the nature of the lipid used, its structure, chemical arrangement, hydrophobicity, physical state, and lipid interactions with the other components of the film (Rhim and Shellhammer, 2005). Lipids are usually combined with other film-forming materials such as proteins or polysaccharides as emulsion particles or multilayer coatings in order to increase the resistance to water penetration (Mehyar *et al.*, 2012).

Polar resin films are good barriers for O₂, CO₂ and ethylene. Hydrophobic substances used for the lipid-based films include natural waxes (carnauba, rice bran and beeswax); petroleum-based waxes (paraffin and polyethylene wax); petroleum-based mineral and vegetable oils (Rhim and Shellhammer, 2005). Wax is the term used for a series of naturally or synthetically produced non-polar substances. Waxes are the efficient barriers to water-vapour transfer due to their hydrophobic nature (Han, 2003).

Protein films

Protein based edible films have received considerable attention in the recent years due

to their ability to be used as edible packaging materials over the synthetic films (Wittaya, 2012). Proteins are superior to other sources like polysaccharide due to their ability to form films with greater mechanical and barrier properties (Cuq *et al.*, 1998). Protein based films are being used as carriers for antimicrobial and antioxidant agents.

Antimicrobial packaging is an emerging technology that could have a significant impact on shelf life extension and food safety. Use of antimicrobial agents in food packaging can control the microbial population and target specific microorganisms to provide higher safety and quality products (Perez-Perez *et al.*, 2006). Protein based films are being used in multilayer food packaging materials together with non-edible films. In this case, the protein based edible films would be the internal layers in direct contact with food materials.

The protein films are generally formed from solutions or dispersions of the protein as the solvent (water, ethanol or mixture) evaporates (Kester and Fennema, 1986). Proteins must be denatured by heat, acid, base and solvent in order to form more extended structures that are required for film formation.

The extended protein chains can associate through hydrogen, ionic, hydrophobic and covalent bonding. Thus protein films are expected to be good oxygen barriers at low relative humidities. Various types of proteins like whey protein, corn Zein, wheat gluten, soy protein, mung bean protein, and peanut protein have been used for film formation (Bourtoom, 2008). Zein has excellent film forming properties and is used for fabrication of biodegradable films.

Su *et al.*, (2007) formulated edible protein films from soy protein isolate through an enzymatic cross-linking method with a purified microbial transglutaminase (MTG)

that was produced from a new effective strain *Streptomyces* sp. WZFF.L-M1 which was followed by addition of glycerol and suitable heating and drying treatments.

The films were about 50 mm thin, had homogenous structures without any observable holes. The films had high water keeping capacity and strong elasticity that the ultimate tensile strength (TS) and elongation at break (Eb) had been increased (TS>5 MPa, Eb>50 %) and that the prevention rates against the permeability of water vapour and oxygen were also upgraded more than 85% and 70 %, respectively.

Wagh *et al.*, (2014) formulated casein and whey protein concentrate (WPC) films plasticized with glycerol and sorbitol. Tensile strength (TS), tensile strain (TE) and elastic modulus (EM) of the films ranged from 0.71 to 4.58 MPa, 19.22 to 66.63 % and 2.05 to 6.93 MPa, respectively. The film properties were influenced by the type of biopolymer (casein and whey protein concentrate), plasticizer and its concentration. The increase in the level of the plasticizer, increased the film thickness, TE and water vapour permeability (WVP), however decreased TS and EM. Casein films showed superior tensile properties as compared to WPC films. The oxygen permeability of casein films was relatively lower than that of WPC films, regardless of the plasticizer used.

Nandane *et al.*, (2015) studied the effect of process parameters on mechanical properties of Soy protein Isolate (SPI) edible film using response surface methodology. The increase in level of SPI increases thickness and tensile strength whereas decrease in young's modulus and elongation at break. Increase in amount of plasticizer, decreased thickness and tensile strength but increased young's modulus and elongation at break. The optimum formulation for meeting the set

criteria of response functions was SPI concentration 8.65%, plasticizer concentration 60%, and pH 8.99.

Bioactive biodegradable packaging

It is defined as the packaging in which the material used is biodegradable like cellophane and the functional additive is of natural origin like nisin (Guerra *et al.*, 2005). According to Lopez-Rubio *et al.*, (2006), bioactive packaging is a way to create healthier packaged foods which have a direct beneficial impact on consumer's health. Bioactive compounds are defined as essential and nonessential compounds that occur in nature and are part of the food chain with some health benefits (Biesalski *et al.*, 2009). The types of bioactive compounds that have been proposed or used in food packaging include enzymes, peptides, polysaccharides, phospholipid analogs, antibodies, oligonucleotides and other antimicrobial agents (Goddard and Hotchkiss, 2007).

Ko *et al.*, (2001) studied the effects of hydrophobicity/ hydrophilicity of edible films against *Listeria monocytogenes* strain V7 by various nisin concentrations (4.0 - 160 IU/film disk) and pH values ranging from 2.0 to 8.0. Mechanical properties and water vapour permeability of films prepared with or without nisin were also compared. Surface hydrophobicities of WPI, SPI, egg albumen and wheat gluten were determined as 446, 282, 232 and 142, respectively. As the nisin concentration increased, the amount of inhibition progressively increased in all tested films. Using nisin, edible films with higher hydrophobicity values of 280 to 450 units under an acidic environment exerted a greater inhibitory effect against *L. monocytogenes*.

Ku *et al.*, (2007) formulated edible films of gelatin and corn Zein by incorporating nisin to the film-forming solutions. Corn Zein film

with nisin of 12,000 IU/ml had an increase of 11.6 MPa in tensile strength compared with the control, whereas gelatin film had a slight increase with the increase of nisin concentration added. Water vapour permeability for both corn Zein and gelatin films decreased with the increase of nisin concentration. Antimicrobial activity against *Listeria monocytogenes* increased with the increase of nisin concentration, resulting in 1.4 log cycle reduction for corn Zein film and 0.6 log cycle reduction for gelatin film at 12,000 IU/ml.

Starch based biodegradable films

Starch is one of the most common and easily obtained natural polymers, making it attractive as a potential bio-based alternative to synthetic polymers. The plasticisation of starch is complex due to the extensive hydrogen bonding between chains (Abdorrezza *et al.*, 2011). This study shows that a simple quaternary ammonium salt combined with hydrogen bond donor (HBD) forms effective modifiers that produce flexible plastics with good mechanical properties that are comparable to some polyolefin plastics. Starch-based plastics can be formed by the same processes as current commercial plastics, giving similar mechanical strength to some polyolefin plastics. The processing conditions are shown to significantly affect the structure of the polymer, which has a concomitant effect upon the mechanical and physical properties of the resulting plastic. Using a glycerol based modifier results in a totally sustainable and biodegradable material which can be formed by extrusion, pressing, vacuum forming and injection moulding. Most significantly, it is shown that these plastics are environmentally compatible, recyclable, bio-degradable and compostable. Starch and cellulose are two of the most abundant polysaccharides, and both are homoglycan polymers. D-glucose is the

monomer unit in both starch and cellulose; however, they have very different mechanical and chemical properties from each other due to a small difference in their structure. Starch is made up mostly of amylose and amylopectin (Avella *et al.*, 2005). The linking oxygen atom is in the axial position, which helps all monomer glucose units to be oriented as each other, indicating that polysaccharide starch is connected by α (1-4) glycosidic linkage, consequently the starch chains interact in a helix. Amylopectin is a branched version of amylose, where α (1-6) glycosidic linkage form a branch. The glycoside linkages begin to breakdown at 150°C while its granules start to decompose above 250°C. A slight degree of reorganisation of hydrogen bonds arises at low temperatures which straightens the polymer chains. The ratios and distributions of amylose and amylopectin vary in each starch depending on its source.

Plasticizers

Plasticizers are non-volatile organic molecules that are added to polymers to reduce brittleness and crystallinity, improve toughness and flexibility, lower glass transition and melting temperatures (Mekonnen *et al.*, 2013). The council of the IUPAC (International Union of Pure and Applied Chemists) defined a plasticizer as “a substance or material incorporated in a formulation (usually a plastic or elastomer) to increase its flexibility, workability, or distensibility”. The compatibility between polymer and plasticizer is a major effective part of plasticization and various parameters including polarity, hydrogen bonding, dielectric constant and solubility parameters (Devlieghere *et al.*, 2004). There are two types of plasticization: internal and external. Internal plasticizers chemically modify a protein chain through addition of substituent group which is attached by covalent bonds.

Internal plasticizers create steric hindrance between the protein chains leading to increased free volume and improved flexibility. External plasticizers solvate and lubricate the protein chains, lowering the glass transition temperature of the proteins and increase the free volume. Common plasticizers used in edible films and coatings are typically polyols including glycerol, propylene glycol, polypropylene glycol, sorbitol and sucrose. Fatty acids have also been used as plasticizers in edible films and coatings. The effectiveness of a plasticizer is dependent upon: size, shape and compatibility with the protein matrix (Sothornvit and Krochta, 2001). McHugh and Krochta (1997) reported that the addition of a plasticizer increased the permeability of a film or coating. The glycerol found naturally in the combined form as glycerides in animal and vegetable fats and oils, is the best plasticizer for water soluble polymers (Muller *et al.*, 2008). The hydroxyl groups present in glycerol are responsible for inter and intramolecular interactions in polymeric chains, providing films with a more flexible structure and adjusting them to the packaging production process (Souza *et al.*, 2012).

Diverse physicochemical properties of plasticizers effect both mechanical and barrier properties of starch films. The addition of amphiphilic plasticizer like fatty acids (palmitic acid and stearic acid) was found to promote the development of polysacchride-lipid sandwich structures in the films.

Essential oils

Essential oils (EOs) are aromatic and volatile oily extracts obtained from aromatic and medicinal plant materials, including flowers, buds, roots, bark, and leaves by means of expression, fermentation, extraction or steam distillation. Approximately 300 EOs are commercially important in the flavour and

fragrance markets Van de Braak and Leijten (1999). Due to their biological properties and flavour characteristics, these oils have been extensively used for centuries in food products. Regarding the meat and meat products, EOs from oregano, rosemary, thyme, clove, balm, ginger, basilica, coriander, marjoram, lemongrass and cinnamon have shown a greater potential to be used as an antimicrobial agent. Besides antibacterial properties (Mourey and Canillac, 2002), EOs or their components have been shown to exhibit antiviral (Bishop, 1995), antimycotic (Mari *et al.*, 2003), antitoxigenic (Juglal *et al.*, 2002), antiparasitic (Pessoa *et al.*, 2002), and insecticidal (Karpouhtsis *et al.*, 1998) properties.

The use of essential oils as natural antimicrobial compounds in foods has attracted growing interest in the recent years, to meet the consumer's requirements in terms of food quality and safety. The antimicrobial activities of different essential oils and essential oil components have already been proved on different species of microorganisms (Di Pasqua *et al.*, 2007), nevertheless, direct incorporation of essential oils in food still encounters technological limitations, related to the hydrophobic, reactive and volatile nature of the bioactive molecules constituting the essential oils (Baratta *et al.*, 1998). EOs have been found to possess potent antibacterial and antifungal activity against several microorganisms associated with meat, including Gram positive and Gram negative bacteria. Generally they consist of more than 60 organic compounds with low molecular weight and large differences in antimicrobial capacity. The major active components of essential oils can be classified in three classes, namely phenols, terpenes, and aldehydes (Ceylan and Fung, 2004). Several works reported that all three classes of components principally act against the cell cytoplasmic membrane (Ceylan and

Fung, 2004), especially because of their hydrophobic nature, which can affect the percentage of unsaturated fatty acid on the membrane and thus alter its structure.

Lemongrass oil

The genus *Cymbopogon* possesses a large number of odoriferous species of the grass family (Poaceae) and is characterized by plants bearing aromatic essential oils in all parts (El-Kamali *et al.*, 1998). All the grasses were first categorized under *Andropogon* and only five species were recognized. Out of total 30 taxa reported from Indian sub-continent, currently we have 21 taxa in India and majority of *Cymbopogon species* can be distinguished from the related genera by the aromatic smell.

Lemongrass is widely used in traditional medicine in many countries. Among its attributable popular properties are those related to analgesic and anti-inflammatory actions (Ortiz *et al.*, 2002). Besides the medicinal use, the lemongrass essential oil is also used in the food (flavouring), perfume and cosmetics industries (Thapa *et al.*, 1971). Lemongrass oil is reported to have potent antimicrobial action against number of organisms. The oil obtained from the *C. citratus* leaves exhibited antimicrobial activity when tested against 42 microorganisms (20 bacteria, 7 yeasts and 15 fungi) (Chalchat *et al.*, 1997). The isolated bacteria presented a superior susceptibility compared to the fungi (Ibrahim, 1992). This plant extracts and/or essential oil, especially the oil for its citral content, presented positive antibacterial activity for *Escherichia coli* (Ogulana *et al.*, 1987). The antioxidant activity of lemongrass oil has also been highlighted in many studies (Baratta *et al.*, 1998) registered that the lemon grass oil had shown anti-oxidizing capacity comparable to that of tocopherol and butylated hydroxyl

toluene (BHT) (Cheah *et al.*, 2001) reported that the dichloromethane and methanolic extracts of this plant showed powerful antioxidant activity.

Cinnamon oil

For thousands of years, cinnamon has been used in spices for culinary uses. Cinnamon (*Cinnamomum zeylanicum* Nees), the evergreen tree of tropical area, a member of family *Lauraceae*, has been used in day to day routine as a spice and condiment in India.

The main compounds isolated and identified in cinnamon (*C. zeylanicum*) belong to two chemical classes: polyphenols and volatile phenols. Among polyphenols, cinnamon contains mainly vanillic, caffeic, gallic, protocatechuic, p-coumaric, and ferulic acids (Muchuweti *et al.*, 2007). The chemical composition of cinnamon oil depends upon the part of plant from which they are extracted. Cinnamaldehyde, is the most represented compound present in bark essential oil ranging from 62-90%, depending upon extraction protocol followed (Ravishankar *et al.*, 2009).

In cinnamon leaf essential oil, the main component is eugenol, which reaches a concentration of more than 80%. Cinnamon oil in general has got good anti-inflammatory, antioxidant, anti-ulcer, anti-microbial, hypoglycemic and hypolipidemic potential. Hili *et al.*, (1997) indicated that cinnamon oils have potential action against various bacteria (*Pseudomonas aeruginosa*, *Staphylococcus aureus*, and *Escherichia coli*) and yeast (*Torulopsis utilis*, *Schizosaccharomyces pombe*, *Candida albicans*, and *Saccharomyces cerevisiae*). Mathew and Abraham (2006) have reported that methanolic extract of Cinnamon contains a number of antioxidant compounds, which can effectively scavenge reactive oxygen species

including superoxide anions and hydroxyl radicals as well as other free radicals under *in vitro* conditions.

Dussault *et al.*, (2014) reported that essential oil obtained from the bark of *C. cassia* can control the growth of the spoilage microorganism *L. monocytogenes* in meat products contaminated at a concentration of 5 ppm, which did not change the sensorial properties of the products. In particular, cinnamon essential oil reduces the bacterial growth rate significantly in artificially contaminated samples when compared with an untreated control. Commercial essential oils obtained from the two most common species of cinnamon, *C. cassia* (leaf-branch) and *C. verum* (bark), were tested against *L. monocytogenes* NCTC 11994, *L. monocytogenes* S0580, *S. typhimurium* ATCC 14028, *S. typhimurium* S0584.

Peppermint oil

Peppermint oil is obtained from the leaves of the perennial herb, *Menthapiperita L.* and *M. arvensis* a member of the Labiatae family. It is a colourless, pale yellow or pale greenish yellow liquid having characteristic odour and taste followed by sensation of cold, freely soluble in ethanol (70%). The oil is found under sides of the leaves, is extracted by steam distillation (Alankar, 2009).

Peppermint oil is commonly used as flavouring in food and beverages and as a fragrance in soaps and cosmetics. Peppermint oil may cause side effects such as heartburn and it may interact with certain medications. Peppermint oil capsules may help to relieve common symptoms of irritable bowel syndrome such as abdominal pain, bloating and gas (Zivanovic, 2005). Peppermint was first described in 1753 by Carl Linnaeus from specimens that had been collected in England; He treated it as a species (Khanna *et al.*,

2014) but it is now universally agreed to be a hybrid (Hoffmann and Lunder, 1984). They are dark green with reddish veins. The leaves and stems are usually slightly fuzzy. Peppermint typically occurs in moist habitats including stream sides and drainage ditches (Neeraj *et al.*, 2013).

Peppermint has high menthol content. The oil also contains menthone and carboxyl esters, particularly menthylacetate (Burt, 2004). Dried peppermint typically has 0.3%-0.4% of volatile oil containing methanol (7%-48%), menthone (20%-46%), menthyl acetate (3%-10%). Peppermint also contains small amount of many additional compounds including limane, pulegone, caryophyllene and pinene. Peppermint contains terpenoids and flavonoids such as eriocitrin, hesperidin and kaempferol 7-O-rutinoside (Sartoratto *et al.*, 2004).

In 2014, world production of peppermint was 92, 296 tonnes, led by Morocco with 92% of the world total reported by FAOSTAT of the United Nations. Peppermint is commonly available as an herbal supplement, there are no established, consistent manufacturing standards for it and some peppermint products may be contaminated with toxic metals or other substituted compounds.

Clove oil

Cloves are a dark brown, aromatic spice that can add an interesting flavor to food and drinks. A chemical called eugenol make up 70 to 90% of the oil and is the chief substance responsible for the aroma and taste of cloves. Cloves are the dried flower buds of the clove tree. The clove tree belongs to the myrtle family of plants. It includes the plants that produce all spice, eucalyptus oil and the bay rum oil that is used in cologne and after shave lotion.

Clove oil has been found to have antimicrobial activity against certain harmful bacteria and yeast in food (Kouidhi *et al.*, 2010). Eugenol is a member of phenylpropanoid class of chemical compounds. It is present in concentration of 80 to 90% in clove bud oil and at 82 to 88% is clove leaf oil (Didry *et al.*, 1994). Eugenol is hepatotoxic, meaning it may cause damage to the liver (Thompson *et al.*, 1998). Overdose is possible, causing a wide range of symptoms from blood in the patient's urine, to diarrhoea, nausea, dizziness on rapid heartbeat (Fujiswa *et al.*, 2002). In context, this would represent a toxic dose in a range of 500-1000 mg/ kg approximately one third that of table salt (Hartnoll *et al.*, 1993). Eugenol is a component of balsam of Peru, to which some people are allergic (Radwan *et al.*, 2014).

For years, many dentists recommended that clove be used at home after dental procedures to disinfect open wounds and to help with any post-surgical discomfort. Today clove oils main use in dentistry is to add flavour to mouth rinse and numbering gels (Barnes *et al.*, 2007). It is also used for its strong antioxidant content, which helps to boost the immune system and fight off infections. It's also great for eliminating fungal infections and alleviating muscle ache pains. By diffusing the oil through the air, air quality can be improved and you may benefit from the mental benefits of clove oil including improved memory, lessened anxiety and improved overall mood. The main chemical components of clove oil are eugenol, eugenol acetate, isoeugenol and caryophyllene. Clove oil is valuable for relieving respiratory problems like bronchitis, asthma and tuberculosis.

The advantage of using edible films is that, they are integral part of the food product. Edible films are in general good moisture barrier, able to inhibit moisture exchange

between food product and atmosphere, preventing microbial growth, texture changes, and undesirable chemical and enzymatic reactions. It also extends the shelf life of food products.

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