Chapter 9
Use of Edible Coatings, a Novel Preservation Method for Nuts

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9.1 Introduction

It is widely known that nuts have high lipid content and are rich in essential fatty acids (especially linoleic and linolenic acids). Their fatty profile very rich in mono- and polyunsaturated fats makes them very prone to lipid oxidation and rancidity. Nonenzymatic oxidation in roasted peanuts is known to be the major cause of rancidity, since high temperature eliminates the activity of lipoxygenase (Lee et al. 2002). In

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their natural state, nuts have natural built-in packaging protection in the form of skins and shells (Miller and Krochta 1997). These natural barriers retain flavor and aroma and regulate the oxygen transport, as well as that of carbon dioxide and moisture.

However, processed foods that lack these natural barriers unfortunately dominate modern dietary choices. Therefore, in minimally processed nuts, the kinetics of lipid oxidation has to be controlled if the development of off-flavors that make the product unacceptable to the consumer is to be controlled. This is of significance because nuts in general and almonds especially are main ingredients of many traditional confectionery products such as *turrón* (a form of nougat) or marzipan, and any degradation of nut quality would consequently lead to loss of product quality.

Nuts are also ingredients in many dishes and widely consumed as snacks. Indeed, nuts are considered a healthy food choice when they form part of a balanced and healthy diet because they have proven cardioprotective effects. According to Abbey et al. (1994), replacing half of the daily fat intake with nuts has been known to lower total and LDL cholesterol levels in humans significantly. Numerous clinical studies have revealed that low-density lipoprotein cholesterol reductions of 10–15 % have been observed, where walnuts, almonds, macadamias, hazelnuts, pecans, or peanuts were incorporated into the diet (Kurlandsky and Stote 2006). Over decades, synthetic plastics (petrochemical based) have been the traditional materials used for the packaging of food products. A variety of synthetic polymers and laminates have been developed and represent an excellent barrier to oxygen transference (Miller and Krochta 1997). The increased use of synthetic packaging films has resulted in serious ecological problems due to their non-biodegradability (Tharanathan 2003). The concern for a safe environment has led to a shift towards the use of biodegradable materials, i.e., edible films and coatings.

9.2 Edible Films and Coatings to Maintain Quality and Safety

In the last few years, the research in the field of edible films and coatings has attracted significant attention within the scientific community. The main advantage of these alternative materials is the reduction of synthetic packaging and the increase of recyclability, while having the potential to limit moisture, aroma, and lipid migration between food components (Miller and Krochta 1997; Nussinovitch 2009).

An edible film or coating consists of a thin layer of an edible material that is applied on the food product and is able to protect it from the environment. When formulating an edible film, at least one of the components must be capable of forming a structural matrix with a sufficient cohesiveness (Debeaufort et al. 1998). This layer should provide a good moisture barrier so various materials such as carbohydrates, proteins, lipids, or hydrocolloids have been suggested and were tested, and found to vary in their suitability. Carbohydrates and proteins were tried but found insufficient due to their hydrophilic nature; however, this issue was mitigated through the addition of lipids to the film formulation, to make composite films (Tharanathan 2003). These multicomponent edible films and coatings have the advantage of possessing the complementary desirable functional properties of each their components and counterbalance any component shortcomings. A composite film formulation can be tailor-made to suit the needs of a specific commodity (Tharanathan 2003). Composite films are generally based on a hydrocolloid structural matrix to which a plasticizer (such as sorbitol or glycerol) is added to promote the formation of edible films and coatings with good mechanical properties. Many materials are too brittle and would not form proper films without the addition of these agents. Brittleness is reduced through the changes in the hydrogen bonding between the film polymers.

An additional advantage of edible films and coatings is that they could act as vehicles for edible active ingredients that could be added to the formulations. The film material could be used to encapsulate selected additives which may have product enhancing or functionality such as an antioxidant action. Other possibilities include the encapsulation of antimicrobial agents, aroma compounds, and pigments, ions that stop browning reactions or nutritional substances such as vitamins (Debeaufort et al. 1998).

The choice of a film-forming substance, and additives, depends mainly on the specific characteristics of the food product to be coated. In general, the most important general prerequisites to be fulfilled by edible films and coatings are the following (Debeaufort et al. 1998):

- Good sensory qualities
- High barrier and mechanical properties
- Enough biochemical, physicochemical, and microbial stability
- Free of toxics and safe for health
- Simple technology
- Nonpolluting
- Low cost of raw materials and process

In the last decade, the research into the use of edible films and coatings to maintain quality and extend the shelf life of food products has attracted significant attention. They have been successfully used in the shelf-life extension of many different products: meat and meat derivatives (Oussalah et al. 2004; Chidanandaiah and Sanyal 2009; Ou et al. 2005), fish and derivatives (Gómez-Estaca et al. 2007; Duan et al 2010; Lopez-Caballero et al. 2005), fruits (Pérez-Gago et al. 2006; Tapia et al. 2008), and vegetables (Ponce et al. 2008; Ayranci and Tunc 2003).

9.3 Application of Tailor-Made Edible Coating to Nuts

Preservation of nuts through the use of corn zein and shellac wax coatings has been used for hundreds of years (Tharanathan 2003). According to Debeaufort et al. (1998), in the nineteenth century, sucrose was initially applied as an edible coating on nuts, almonds, and hazelnuts to prevent oxidation and rancidity during storage. In recent years, a variety of materials have been tested in order to meet the specific requirements of this type of products. In the search for suitable film-forming materials for the proper coating of nuts, the following requisites should be taken into account.

9.3.1 Organoleptic Compatibility

Being considered as food components, edible films and coatings are usually required to be as tasteless as possible in order not to be detected during the consumption (Contreras-Medellin and Labuza 1981). When applied to nuts, edible coatings should not modify the taste and flavor of the product. If this is not possible, the organoleptic properties of the coating should be compatible with that of the nut (Biquet and Labuza 1988).

9.3.2 Antioxidant Activity

Given that nuts are rich in oxidation-sensitive lipids, the coating material should be able to protect these components from the oxidation process that would result in off-flavors. Edible films and coatings that are able to do so, are claimed to have antioxidant activity. This protective capability of the edible films and coatings is the result of two different actions:

(a) Barrier to oxygen. Among all the factors affecting the rate of lipid oxidation, oxygen concentration is one of the most important (Labuza 1971). If oxygen availability is reduced by the barrier action of the coating, the oxidation rate is diminished. For this reason, materials with low oxygen permeability are the best choice in order to obtain the best coatings for oxidation-sensitive products such as nuts (Nussinovitch 2009; Swenson et al. 1953). In this respect, ambient relative humidity plays an important role in the stability of the product because the oxygen permeability of edible films and coatings depends greatly on water availability. The greater the relative humidity, the greater the water content in the film, which promotes molecular mobility and diffusion controlled processes, such as oxygen permeation (Hong and Krochta 2006).
(b) Specific action of antioxidant additives. As commented on above, edible films and coatings are able to encapsulate active ingredients or additives exhibiting some specific action in the system. The incorporation of antioxidants is recommended in our context (Nussinovitch 2009; Cosler 1957), given that these chemicals will have an antioxidant action on the product. In the last few years, natural antioxidants have received a great deal of attention because of the worldwide trend to avoid synthetic food additives. According to Frankel (1996), natural antioxidants in food products may have clear benefits because they have anticarcinogenic effects and inhibit biologically harmful oxidation reactions in the body. Due to their antioxidant and antimicrobial properties, essential oils are being studied as additives to be incorporated into edible films and coatings (Atarés et al. 2010).

9.3.3 Good Adhesiveness on the Surface of the Nut

The antioxidant effect of the coating will be effective so long as the coating is in close contact with the nut surface. Evidently, the occurrence of cracks and flaking in the coating would drastically affect its barrier properties. Generally speaking, the adhesiveness of the coating material on the food product is highly dependent on the nature of both, as well as their affinity to each other. Adhesiveness depends on the nature and on the number of interactions between film and support (Debeaufort et al. 1998).

In the case of nuts, the inherently poor adherence of coating (hydrophilic) and nut surface (hydrophobic) may be the cause of incomplete coverage, which is a common and important setback. According to Lin and Krochta (2005), this problem could arise during the coating step (dewetting), the drying (shrinking and cracking), and the storage of the coated product (flaking). The addition of a surfactant (aka emulsifiers or surface active agents) to the coating formulation is a common technique to solve this problem. They allow adherence and mixing between hydrophobic and hydrophilic materials. Their incorporation would reduce the interface tension between coating and solid, hence the interactions between both are promoted and the extensibility is improved. An alternative solution would imply the modification of the nut surface with surfactant adsorption prior to the coating. Both techniques will be commented on in Sect. 9.4.

9.3.4 Microbial Stability

Contamination of nuts by aflatoxins is a serious health problem. These toxins are produced from infection by fungus *Aspergillus* and are considered as one of the most dangerous contaminants of foods (Basaran et al. 2008; Luttfullah and Hussain 2011). The incorporation of an antimicrobial agent should also be considered in the formulation of coatings for nuts.

9.3.5 Good Optical Properties

An additional interesting requisite for nut coating materials would be their ability to improve the general appearance of the coated product. In this respect, the color of the nuts should not be altered and the coatings should exhibit high transparency. The gloss of the nuts can be improved to make them more appealing. Water-soluble cellulose derivatives (methylcellulose (MC), hydroxypropyl cellulose (HPC), carboxymethyl cellulose (CMC), and hydroxypropyl methylcellulose (HPMC)) produce transparent and shiny films (Debeaufort et al. 1998).

Table 9.1 Original research studies—performed over the last two decades on the application of edible coatings to nuts

Nut product	Coating		Reference
Peanuts	Whey protein	Sugar esters, soy lecithin, Sorbitan laureate (Span 20) (*s*)	Lin and Krochta (2005)
		Glycerol (*p*)	Lee et al. (2002)
		Lecithin (*s*)	
		Methyl paraben (*am*)	
		Vitamin E (*ao*)	Lee and Krochta (2002)
		Glycerol (*p*)	Maté et al. (1996)
		Distilled acetylated monoglycerides	
		Glycerol (*p*)	Maté and Krochta (1998)
Pecans (pecan kernels)	Methyl cellulose (MC), hydroxypropyl cellulose (HPC), carboxymethyl cellulose (CMC)	Propylene glycol (PG), sorbitol (*p*)	Baldwin and Wood (2006)
		Lecithin (*s*)	
		α-tocopherol (vitamin A), butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT) (*ao*)	
Almonds	Hydroxypropyl methylcellulose (HPMC)	Tween 80 (*s*)	Atarés et al. (2011)
		Ascorbic acid, Citric acid, Ginger oil (*ao*)	

p plasticizer, *s* surfactant, *am* antimicrobial, *ao* antioxidant

9.4 Edible Coatings for the Extension of Shelf Life

Other than maintaining organoleptic and other quality attributes, the latest advances in the application of edible coatings have been developed to extend the shelf life of nuts. A number of studies on the oxygen and aroma barrier properties of edible films have been published between 1967 and 2005 presenting varied potential film models (Miller and Krochta 1997). Later studies (Table 9.1) show that suitable film-forming materials include whey protein isolate and cellulosic derivatives which are all good barriers to oxygen (Han and Krochta 2007; Maté and Krochta 1998; Lee and Krochta 2002; Trezza and Krochta 2002; Miller and Krochta 1997), which make them suitable for this use. The same studies state that these materials are good barriers to aroma and oil.

Lin and Krochta (2005) dealt with the adherence of whey protein coatings on peanuts which is vital for shelf-life extension. Peanuts were immersed in aqueous solutions of the surfactants and air dried to modify the peanuts' surface energy which rendered it more compatible with the hydrophilic coating. The addition of surfactants (Span 20) to the whey protein coating solution also improved the coverage of the peanuts in a concentration-dependent fashion Maté et al. (1996) investigated humidity in peanuts and its effect on coatings, and concluded that the factors determining the effectiveness of the coatings were their thickness and the relative humidity, which indicated that the mechanism of protection was due to oxygen barrier properties. The lack of discontinuities of the coatings was critical in improving the shelf life of peanuts.

Protein coatings have been found to be beneficial in that whey protein coatings delayed oxygen uptake and rancidity of dry roasted peanuts when compared to the uncoated controls. Maté and Krochta (1998) found that coated peanuts were definitely more resistant to oxidation than those uncoated and hexanal levels which is a degradation product of linoleic acid and an indicator of lipid oxidation and therefore rancidity was found to be lower (Lee et al. 2002; Lee and Krochta 2002) confirm this fact. This led to the conclusion that coated samples were oxidized significantly slower than the uncoated reference, meaning a shelf-life extension of coated samples.

Other studies such as that of Baldwin and Wood (2006) investigated the application of cellulosic edible coatings on pecan kernels using two plasticizers (propylene glycol and sorbitol), lecithin as surfactant, and several antioxidants (vitamin E, BHA, and BHT). The coatings imparted gloss to the nuts. Determination of degradation products (hexanal) by gas chromatography revealed that hexanal levels were at least twice as low in coated kernels as in uncoated controls, meaning that coated kernels underwent less fat oxidation and were less rancid, which correlated with the sensory analysis, thus further proving the validity of selected coating materials for the preservation of quality and product shelf-life extension.

9.5 Application of Edible Coatings to Almonds

Almonds are one of the most commonly used nuts and are a highly nutritional source of vitamins (B complex) and minerals (Mg, P, K) (Ahmad 2010). They contain high levels of fat (51 % w/w), but have a favorable fatty acid profile (64–82 % oleic acid, 8–28 % linoleic acid, 6–8 % palmitic acid). Here too, the oxidation process of unsaturated fatty acids can produce unpleasant rancid off-flavors, thus shortening the shelf life of almonds under ambient conditions. Almonds with hydroxypropyl methylcellulose (HPMC) formulations containing different antioxidant additives such as ascorbic and citric acids, have been frequently studied. Other materials which may contribute to the organoleptic properties of the nut are potential additives to coating materials. Atarés et al. (2011) added ginger essential oil into HPMC coatings to be applied to almonds, and studied the oxygen permeability of the films in order to characterize the mechanisms that modulated the lipid oxidation protection. Results in Table 9.2 show that acids incorporation produced a significant improvement of the oxygen barrier performance of the films. However, the addition of ginger oil had opposite negative effect on film integrity. This led to the conclusion that whenever materials were added to coating systems they had to be tested rigorously as any material added may affect the coating efficacy.

9.6 Conclusion

Edible films and coatings are a promising alternative to conventional packaging systems that among their functions can be added maintenance of organoleptic and other quality attributes. Many functions of edible packaging are identical to those of plastic films. However, their use would still require an overpackaging, notably for handling and hygiene purposes (Debeaufort et al. 1998).

The use of a coating acting as an oxygen barrier combined with a simpler plastic film acting as a moisture barrier represents an alternative packaging system. The film-forming techniques are critical for the performance of the films (Maté and Krochta 1998) and more work is needed to improve the efficiency of the coatings and achieve longer rancidity delay with thinner films.

Microbial stability will become more and more important as more edible polymers approach commercial availability (Miller and Krochta 1997). And its effect on other properties of the films and the product (oxygen permeability, organoleptic properties, and others) also needs further investigation.

The mechanism of action of each specific compound or mixture should be considered individually to get the best match of the coating formulation and the requirements of the product. Such studies and the exploitation of these results have impact on the quality of nuts in general.

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