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Review

Essential oils as additives in biodegradable films and coatings for active food packaging

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ABSTRACT

Background: Petroleum derivate plastics represent a serious environmental problem, which is why alternative sustainable solutions must be found. To this aim, recent research has focused on the development of edible/biodegradable packaging for food products. The implementation of this novel packaging requires analyzing thoroughly the effect of the ingredients used on the most relevant properties of the material.

Scope and approach: Essential oils represent an interesting ingredient for biodegradable food packaging, mainly due to their natural origin and their functional (antioxidant/antimicrobial) properties, allowing for obtaining active materials aiming to extend shelf-life and add value to the product. However, their inclusion in edible/biodegradable films for food packaging may imply some impact on several properties of the system (such as optical, tensile…), affecting in turn the consumer acceptability. Before the increasing research on biodegradable materials for food packaging, and the growing interest on natural food additives, this paper aims to review the latest findings on how essential oils impact the most relevant properties of edible films and coatings, namely microstructural, physical (tensile, barrier, optical), antioxidant and antimicrobial.

Key findings and conclusions: Essential oil incorporation affects the continuity of the polymer matrix, leading to physical changes depending on the specific polymer-oil components interactions. Generally, the film structure is weakened by the oil addition, whereas the water barrier properties are improved and the transparency is reduced. Essential oils may provide the films with antioxidant and/or antimicrobial properties. The oil composition and the specific interactions with the polymer determine its effectiveness as an active ingredient.

1. Introduction

Conventional plastics are derived from petroleum, which entails serious environmental concerns. Biodegradable films and coatings represent an interesting alternative to conventional plastic materials, which is why several biopolymers have been exploited to develop materials for eco-friendly food packaging (Azeredo, 2009). Edible films are thin layers of edible materials, which once formed can be placed on – or between-food components, whereas an edible coating is formed as a coating on a food product. The most common materials for the formulation of edible/biodegradable films and coatings are polysaccharides, proteins and lipids, and the combination of these allows for producing blends of improved characteristics (Fabra, Jimenez, Atarés, Talens, & Chiralt, 2009).

Aiming to the reduction of the use of chemical additives in food industry, growing interest has risen recently on the use of natural food additives with antimicrobial and antioxidant properties that do not have any negative effects on the human health (Alves-Silva et al., 2013). Essential oils (EOs) extracted from plants and spices exhibit antimicrobial and antioxidant properties (Viuda-Martos et al., 2010), which makes them interesting additives in food industry. In addition, most of them are classified as *Generally Recognized as Safe* (GRAS) (Ruiz-Navajas, Viuda-Martos, Sendra, Perez-Alvarez, & Fernández-López, 2013). However, their use as food preservatives is often limited due to their strong flavor. In order to avoid this problem, EOs can be incorporated into edible films (Ruiz-Navajas et al., 2013).

In the recent years, EOs have been extensively studied as additives in edible/biodegradable emulsified films and coatings. Due to their lipidic nature, they are expected to help reduce the water

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vapour permeability of hydrophilic films. Moreover, they have proved to have some impact on other film properties (tensile, optical, structural…), as well as providing antioxidant and/or antimicrobial effects. This paper reviews the role of EOs as additives in edible/biodegradable films and coatings for food packaging, namely their effect on the (1) microstructural and physical properties, (2) antioxidant power and (3) antimicrobial capacity of the films. Recent studies dealing with the characterization and/or application of edible films and coatings incorporated with EOs have been systematically classified, and their main conclusions summarized, in order to give an overview on the state of the art.

2. Effect of EO incorporation on the film microstructure

Table 1 shows a representative selection of recent studies (2005–2015), giving an overview of the tests usually performed when dealing with the effect of EOs addition on the structure and physical properties of edible films.

The qualitative observation of the components arrangement into the film structure is often accomplished using Scanning Electron Microscopy (SEM) or Transmission Electron Microscopy. SEM has been repeatedly utilized to investigate the structure of edible films incorporated with EOs, often in comparison to the film with no lipid.

Contrarily to food packaging materials based on non-polar conventional plastics, edible biodegradable films are usually based on hydrophilic materials such as proteins or polysaccharides, and films or coatings are mainly obtained by casting of the film forming aqueous dispersions and subsequent drying. The incorporation of essential oils in the film forming dispersion is carried out by applying emulsification or homogenization techniques, where fine emulsions of essential oils are obtained containing polymer at the continuous aqueous phase. In dried films, lipid droplets remain embedded into the polymer matrix, as can be observed by microscopic techniques.

It is normally observed that the final microstructure of the films is influenced by the structural arrangement of the components in the film forming dispersion. Moreover, their development during the drying period plays an important role, given that destabilization phenomena such as droplet flocculation, coalescence and creaming can occur. In line with water evaporation, the concentration of the oil dispersed phase increases, thus promoting droplet flocculation rate. Subsequently, coalescence and creaming lead the droplets to the forming film surface, where compounds evaporation occurs together with water (steam distillation). In fact, losses of EOs were reported depending on the polymer forming film (Sánchez-González et al. 2011a, b). Factors promoting the emulsion stability, such as the polymer adsorption at the droplet surface and viscosity increase of the continuous phase, contribute to limit the losses of EOs and affect the final film microstructure. So, polymer-

Table 1
Recent papers studying the effect of essential oils on films properties. (HPMC: hydroxyproplymethylcellulose; RT: room temperature).

	Matrix	Essential oils	Matrix: oil proportion	T-%RH conditioning	Physical & microstructural tests								Reference
					SEM	AFM	Tensile	WVP	OP	Colour	Opacity	Gloss	
Polysaccharides	Lignocellulose	Cedarwood	1:0–0.2	23°C-50%	−	−	+	+	−	−	−	−	Shen and Kamdem (2015)
	HPMC	Tea tree	5%:0.5–2%	20°C-54.4%	+	−	+	+	−	+	+	+	Sánchez-González et al. (2009)
	HPMC	Ginger	1:0.1	25°C-33/53%	+	−	+	+	+	+	+	+	Atarés et al. (2011)
	HPMC, chitosan	Bergamot, lemon, tea tree	1:0.5–2	20°C-54.4%	−	−	+	+	−	−	+	+	Sánchez-González et al. (2011a)
	Chitosan	Cinnamon	2%(w/v):0.4–2% (v/v)	25°C-51%	+	−	+	+	−	+	−	−	Ojagh et al. (2010)
	Chitosan	*Zataria multiflora* Boiss, grape seed	20:0–20	25°C-52%	−	−	+	+	−	+	−	−	Moradi et al. (2012)
	Chitosan	Bergamot	1:0–3	20°C-54.4%	−	−	+	+	−	−	+	+	Sánchez González et al. (2010)
	Chitosan	Green tea extract	2%: 0–20% (w/v)	25°C-50%	−	−	+	+	−	+	+	−	Siripatrawan and Harte (2010)
	Chitosan	Basil, thyme	1:0.5–1	5°C-58%	+	−	+	+	−	−	−	+	Bonilla et al. (2012b)
	Alginate	Garlic	1 g:0–0.4 ml	–	−	−	+	+	−	+	−	−	Pranoto et al. (2005)
	Alginate	Oregano	1:0–1	25°C-50%	+	−	+	+	−	+	−	−	Benavides et al. (2012)
	Alginate	Ginseng	2%:0.5 g/ml	–	+	−	+	+	−	−	+	−	Norajit et al. (2010)
	Alginate	Thyme, lemongrass, sage	3%:1%	25°C-50%	+	−	+	+	−	+	+	−	Acevedo-Fani et al. (2015)
	κ-carrageenan	*Satureja hortensis*	1% (w/v): 1–3% (v/v)	25°C-53%	+	+	+	+	−	+	+	+	Shojaee-Aliabadi et al. (2013)
Proteins	Fish gelatin and chitosan	*Origanum vulgare*	1.75%:0.4–1.2%	25°C-50%	+	+	+	+	−	−	+	−	Hosseini et al. (2015)
	Soy protein	Cinnamon, ginger	1:0.025–0.100	25°C-33%	−	+	+	+	−	+	+	+	Atarés et al. (2010a)
	Na caseinate	Cinnamon, ginger	1:0.025,0.075	25°C-33/53%	+	+	+	+	−	+	+	+	Atarés et al. (2010b)
	Fish protein	Clove, garlic, origanum	4 mg:1 μl	RT-57%	−	−	+	+	−	+	+	−	Teixeira et al. (2014)
	Fish skin gelatin	Bergamot, kaffir lime, lemon, lime	1:0.5	25°C-50%	+	−	+	+	−	+	+	−	Tongnuanchan et al. (2012)
	Gelatin	Bergamot, lemongrass	3:0–20	25°C-50%	+	−	+	+	−	+	+	−	Ahmad et al. (2012)
	Gelatin	Oregano, lavender	1:0.04–0.12	25°C-65%	+	−	+	+	−	+	−	−	Martucci, Gende, Neira, and Ruseckaite (2015)
	Hake protein	Thyme	1 g: 0.025–0.25 ml	RT-57%	−	−	+	+	−	+	+	−	Pires et al. (2011)
	Hake protein	Citronella, coriander, tarragon, thyme	1 g: 0.25 ml	RT-57%	−	−	+	+	−	+	+	−	Pires et al. (2013)
	Whey protein	Oregano	1:0.1–0.3	25°C-53%	−	−	+	+	−	−	−	−	Zinoviadou et al. (2009)