Essential Oils Stabilization into Biodegradable Food Packaging Materials

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Human health, food spoilage, and plastic waste, which are three great topical concerns, intersect in the field of food packaging. This has created a trend to replace synthetic food preservatives with natural ones, to produce bio-functional food packaging, and to shift towards biodegradable polymeric materials. Essential oils are gaining more and more attention in food packaging applications due to their various benefits and fewer side-effects. Fixation into polymeric matrices by emulsification and electrospinning, represents suitable strategies for protection and stabilization of essential oils, promoting the benefits and reducing the drawbacks. This review focuses on creating correlations between the use of essential oils as natural additives, stabilization methods, and biodegradable polymeric matrices or substrates in developing bioactive food packaging materials.

Keywords: bioactive packaging ; herbal extracts ; food safety

1. Introduction

Essential oils are plant secondary metabolites which are thought to be safer than synthetic chemical preservatives. They are generally recognized as safe (GRAS) and approved by the U.S. Food and Drug Administration (FDA), therefore suitable for use in food-related applications without the need for approval. Essential oils usually have complex and particular composition that allows them to act in different ways on a broad range of food pathogens, e.g., Gram positive (G+) or Gram negative (G−) bacteria, molds, etc.

The overall aim of the incorporation/encapsulation strategies in active packaging is to ensure the availability for a large period of time of a greater amount of the bioactive compounds in the final material. Since natural compounds have large variation in composition and properties, one cannot establish general stabilization methods that could suit a broad spectrum of materials. The selection of an incorporation method usually is particular, depending both on the properties of the bioactive components and of the
embedding materials, and also on the targeted food.

The ***incorporation of essential oils into packaging materials*** can be usually performed by (*i*) direct addition to the polymeric materials in earlier steps of material preparation; (*ii*) adsorption or coating onto support materials; (*iii*) immobilization onto the surface of the packaging, (*iv*) mechanical entrapment into physical carriers as accessories for food packaging; (*v*) introduction into headspace; (*vi*) microencapsulation in carriers followed by incorporation into matrices.

2. Emulsion Techniques

Emulsion techniques have owned over time an important place in the food industry but lately gained tremendous attention in active food packaging as an encapsulating method. An emulsion represents a colloidal system consisting of at least two immiscible liquids; the oil, which is a property of main interest in food packaging materials, and the aqueous phase, wherein one liquid (the continuous phase) includes a dispersion of droplets of the other liquid (the dispersed phase). The emulsion technique is used in food packaging mainly as a loading method for the development of active materials in the form of capsules, films, coatings, and emulsified gels. Thus, the obtained materials aim, besides the abovementioned functions, to ensure the release of loaded bioactive components in a controlled manner. Currently there is a growing interest in the development of active food packaging materials based on renewable and environmentally friendly polymers. In the field of bioactive materials containing sensitive essential oils as natural active components, the water soluble polymers have become promising candidates for replacing solvent-soluble and thermal processed materials, since natural bioactive oils have high volatility or are prone to thermal degradation. In active food packaging applications, the emulsion casting films are the most common. Until now there has been no study that demonstrates stabilization of an emulsion solely by biopolymer presence, therefore combination with surfactants or additional processing treatments is applied. To achieve a better protection of sensitive bioactive oils, a pre-encapsulation stage is often involved, which mainly implies loading oil into capsules. Current evidence on the topic of active food packaging based on emulsions shows that the main interest is directed towards the development of new compositions and few researches have also considered technology tuning.

3. Electrospinning

Electrospinning is a non-mechanical processing technique that uses a high-voltage electrostatic field to electrohydrodynamically stretch droplets from melts and especially from solutions into single, continuous, and very fine jet, at nanometric up to several micrometers scale, which solidifies as self-assembling threads to form nonwoven mats of entangled long fibers. This is a simple, versatile, flexible, cost effective continuous process that can be easily upgraded to industrial scale for production of nanofibers with controlled diameter and tailored properties. Since the electrospinning process is conducted at room temperature, without the need of heating usually involved in the preparation of classical packaging materials, it can be used for inclusion and protection of volatile or thermally sensitive additives, such as essential oils or their components. The high surface area to volume ratio of nanofibers makes them sensitive to external factors; for example, temperature, relative humidity, and pH. Application of electrospinning in food packaging is very limited compared with the strongly increasing use of this technique in various other domains. One constraint comes from the necessity of using food grade polymers that should be GRAS or FDA-approved. Besides controlling the viscosity and surface tension of the solution, and the morphology of polymeric macromolecular chains, solvents and their volatility also have an important role in the porosity of electrospun fibers. Bioactive components can be loaded into the porous structure of the nanofibers for protection and controlled release.

Electrospinning is increasingly used for encapsulation of essential and vegetal oils or their principal constituents into nanofibers of biodegradable polymers for use as food packaging materials. Most reports considered direct addition of essential oils into polymeric systems before electrospinning, while in some works only the polymeric materials were electrospun to obtain nanofibers or coatings that were afterwards loaded with the low molecular bioactive principle. While direct addition has the advantage of being a one-step process and of incorporating the entire amount of the bioactive compound into the polymeric system, this might affect the electrospinning behavior or the properties of obtained materials, especially when various solvents are needed; for example, when there is a difference in the hydrophobic/hydrophilic nature of the additive/polymeric matrix. On the other hand, loading the bioactive compound after the electrospinning process requires higher amounts of reagents and additional processing steps but has lower impact on the physical properties of the material. Some other studies considered carriers for encapsulation and protection of active agents inside the polymeric matrix. The carrier system including the active principle can be obtained by electrospinning/electrospraying then added to the polymeric matrix. Post treatments such as conditioning, chemical crosslinking and physical welding or annealing, were used in several studies to improve the properties of the electrospun fibers. These can induce crystallization of components, provide better adhesion of electrospun fibers onto base film, slow the release of the active compounds or convert the electrospun fiber mats into films.

In the case of packaging systems obtained by emulsification, a unified trend was noted in preparation procedures, namely most research has involved the use of Tween 80 as surfactant, glycerol as plasticizer, and ultrasonication to lower the droplet size. The polymeric matrices were mainly aqueous soluble polysaccharides or proteins, hence direct oil-to-water emulsions were obtained, and very few works considered inverse emulsions. Particular and more efficient systems were developed when using solid-particle stabilizers (Pickering emulsions) instead of classical surfactants. Electrospinning was largely used as a versatile method that allows different approaches for fixation of essential oils, such as direct addition into polymeric systems, loading on electrospun fibers, and encapsulation into carriers before electrospinning. Post treatments were used to improve the performance of fibers or to convert fibrous mats into films. Electrospinning enhanced the bioactivity and induced the controlled release of essential oils due to nanostructuration and increased surface to volume ratio. Research was generally focused on varying the components and their concentration in studied systems and less on finely tuning the process parameters. Antimicrobial performance of materials was tested mainly in vitro on isolated pathogens, especially on *E. coli* and *S. aureus* bacterial strains, but overall, relatively few studies involved tests on food.

The use of essential oils in the field of active food packaging is still in its incipient stage, far from the necessary applicability on a larger scale and even further away from the application at industrial level.