



## Use of the electrohydrodynamic process to develop active/bioactive bilayer films for food packaging applications



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### ABSTRACT

In this work, the electro-hydrodynamic process was used to encapsulate an antioxidant, alpha-tocopherol, using different hydrocolloid matrices (whey protein isolate–WPI-, zein and soy protein isolate–SPI-) as shell materials. These hybrid structures were directly electrospun/electrosprayed as a coating layer onto one side of a thermoplastic wheat gluten film, thus giving rise to active/bioactive bilayer packaging structures. The water vapour barrier efficiency of thermoplastic wheat gluten films were improved by the presence of the active coating layer. Moreover, when submitting these structures to a typical industrial sterilization process, the alpha-tocopherol stability was preserved, especially when zein was used as shell material. The amount of the alpha-tocopherol released in the aqueous media was assessed by a spectrophotometric method. The alpha-tocopherol release profiles showed a delayed release of the encapsulated antioxidant in zein matrices (fibres morphology). The release was faster for WPI and SPI-GG probably due to both the capsule morphology and the swelling and faster dissolution of these water-soluble protein matrices. This work provides a new method for developing active/bioactive packaging systems of interest in food applications.

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### 1. Introduction

There is a current trend to incorporate, into packaging materials, active agents that will maintain and enhance the quality and safety of packaged foods. Specifically, antioxidant active packaging seeks to prevent or slow down the oxidation of certain food components, like lipids and proteins, which lead to the deterioration of physical characteristics (such as flavour and colour) of those food products. This active material approach requires the intentional incorporation of antioxidants within the packaging materials and their further migration to those foods (Robertson, 2012).

Polymers, and in particular biomass-derived polymers, are the preferred materials for active packaging because of their intrinsic properties, constituting an ideal carrier for active agents, with the advantage of being adaptable in terms of controlled release and the possibility of combining several polymers through blending or multilayer assembly in order to tailor the application. The drawbacks that initially characterized these biopolymers in terms of poor barrier properties and high instability have, in turn, resulted in

novel applications, making highly permeable and water-plasticizable biopolymers an ideal partner for active packaging where the package is no longer a passive barrier, but actively contributes to the preservation of food by controlled release of the substances (Martínez-Abad, Ocio, & Lagaron, 2014).

However, many active agents are thermally sensitive and, thus, cannot be directly incorporated during typical processing methods used for polymeric materials. This is, for instance, the case of essential oils which have been widely incorporated in diverse biopolymeric matrices for active packaging development due to their antioxidant and antimicrobial effects. However, their high volatility results in the evaporation of most of the active agent during film formation, even when casting methods were employed (Sanchez-Silva et al., 2014).

In a recent work (Fabra, Jiménez, Talens, & Chiralt, 2014), it was demonstrated that the antioxidant capacity of alpha-tocopherol was reduced upon incorporation within pea protein and sodium caseinate matrices by the solvent cast methodology. This reduction ranged from 20% to 30% with respect to the theoretical values initially added to the film and these results were attributed to the loss of alpha-tocopherol activity during the film-formation stages. At an industrial level, the usual thermal processing of bioplastics make the incorporation of active compounds necessary during melt

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compounding or other heat involving steps which would imply some losses of the antioxidant compounds due their thermo-sensitive nature. For instance, Wook et al. (2013) reported losses of resveratrol and  $\alpha$ -tocopherol incorporated in PLA/starch blend films during the polymer melt blending, ranging between 4 and 26%, depending on the film formulation.

Therefore, in an effort to reduce the losses in antioxidant activity during film formation or packaging structure development, the electrospun fibre encapsulation of alpha-tocopherol is being here proposed as a feasible route to stabilize and dose the active agent into biopackaging elements. Aside from the conventional micro-encapsulation techniques, such as spray drying or coacervation, the electro-hydrodynamic process (also called electrospinning) has been recently suggested as being a simple and straightforward method of generating submicron encapsulation structures for a variety of bioactive molecules (Bakhshi, Nangrejo, Stride, & Edirisinghe, 2013; Bock, Dargaville, & Woodruff, 2012; Fernandez, Torres-Giner, & Lagaron, 2009; López-Rubio & Lagaron, 2012; Pérez-Masiá, Fabra, Lagaron, & López-Rubio, 2013; Torres-Giner, Gimenez, & Lagaron, 2008; Xie, Li, & Xia, 2008) and also to generate fibre based interlayer packaging elements (see below). This technique makes use of electrostatic forces to produce electrically charged jets from viscoelastic polymer solutions which upon drying, by the evaporation of the solvent, give rise to ultrathin structures (Li & Xia, 2004). By using the electro-hydrodynamic process already described for the development of multilayer biodegradable systems (Fabra, López-Rubio, & Lagaron, 2014; Fabra, López-Rubio, & Lagaron, 2015), active food packaging materials can be developed by encapsulating hydrosoluble and/or liposoluble active compounds into biodegradable matrices. This technology offers several advantages in protecting the active compound encapsulated into hydrocolloid matrices (Pérez-Masiá, Lagaron, & López-Rubio, 2014). Furthermore, the use of electrospinning is crucial for attaining good adhesion between the hydrophilic natural polymers, such as proteins and the hydrophobic layers (both immiscible), as it has been reported by Fabra et al., 2015. By using this cutting edge approach now described, new active packaging can be formulated in just one step with the additional advantage of simultaneously and intrinsically producing both interlayers with encapsulation performance. The antioxidant activity of the active compound (which could be applied within the multilayers) could be preserved by the encapsulation process induced by the electro-hydrodynamic process.

This proof-of-concept study should provide a new route to explore further for the development of novel multilayer systems with an active role in food packaging. Therefore, this work is aimed at evaluating the availability of the electro-hydrodynamic process in order to develop bilayer active packaging systems with antioxidant properties. To this end, tocopherol was used as a model antioxidant to develop nanostructured coatings and the active bilayer films were characterized in terms of physico-chemical properties, antioxidant activity and release kinetics in water. The stability of the alpha-tocopherol incorporated within the packaging structures was assessed when exposed to a typical industrial sterilization process.

## 2. Materials and methods

### 2.1. Materials

Whey protein isolate (WPI) was kindly donated by ARLA (ARLA Food Ingredients, Viby, Denmark). Soy protein isolate (SPI) was donated by The Solae Company (Switzerland). Guar gum was purchased at Capers Community Markets (Canada). Glycerol was provided by Panreac Quimica, S.A. (Barcelona, Spain). Wheat gluten

(WG), zein from corn (grade Z3625, 22–24 kDa), Tween 20, Tween 80 and alpha-tocopherol (T) (>95.5% of purity) were provided by Sigma–Aldrich Chemie GmbH (Steinheim, Germany) and they were used as received, without further purification.

### 2.2. Film preparation

#### 2.2.1. Development of thermoplastic wheat gluten films (TPWG)

Plasticized wheat gluten films were obtained following the methodology previously described by Chen, Reddy, Wu, and Yang (2012). Wheat gluten was thoroughly mixed with 20% (w/w) glycerol and the mixture was then spread evenly on Teflon and placed in a compression mould (Carver 4122, USA) at a pressure of 38 kPa at 100 °C during 10 min.

#### 2.2.2. Development of nanostructured multilayer WG-based systems

**2.2.2.1. Preparation of the hydrocolloidal dispersions.** The WPI and SPI-GG dispersions were prepared as previously described in Pérez-Masiá et al., 2014. Briefly, 10 wt.-% and 30 wt.-% of the SPI and WPI were dispersed in distilled water, respectively. 1 wt.-% of guar gum and 5 wt.-% of Tween 20 were added to the SPI aqueous dispersion while 10 wt.-% of glycerol and 5 wt.-% of Tween 80 with respect to the wheat gluten weight were dispersed into the WPI solution. The zein electrospinning solution was prepared as described in Fabra and López-Rubio et al., 2014. To this end, 33 wt.-% of the protein was dispersed in a solution of ethanol/water 85:15 (v/v). Afterwards, alpha-tocopherol (50 wt.-% respect to the polymer weight) was incorporated and stirred for 30 min pH of the electrospinning solutions was measured before the electrospinning process and it was ranged between 6.4 and 7.0 in all cases.

**2.2.2.2. Development of the nanostructured coatings.** Thermoplastic WG films were coated with the nanostructured WPI, SPI–GG or zein mats, with or without the antioxidant, produced by means of the electro-hydrodynamic technique. The hydrocolloid mats were directly electrospun/electrosprayed onto one face of the plasticized wheat gluten films using the Fluidnatek™ LE10 laboratory tool of Bioinicia (Paterna, Spain) equipped with a variable high voltage 0–30 kV power supply and a horizontal flat collector where the thermoplastic WG films were located (Fabra et al., 2015). The biopolymer dispersions were introduced in 5 mL plastic syringes and were electrosprayed/electrospun under a steady flow rate using a stainless steel needle with internal diameter of 0.9 mm. The needle was connected through a PTFE wire to the syringe. The syringe was lying on a digitally controlled syringe pump while the needle was in horizontal towards a stainless steel plate attached to a copper grid used as collector and the solvent was vented out of the chamber during the electrospinning run. The experiment was carried out at ambient conditions (20 °C and 40% RH). The distance between the needle and the collector was 10 cm and the voltage was varied from 10 to 15 kV. The flow rate was varied from 0.15 to 0.3 mL/h. Specifically, the flow rate was set at 0.15 mL/h for WPI and SPI-GG dispersions and 0.30 mL/h for the zein electrospinning solutions. The total amount of the electrospun coatings ( $\sim 1.15 \pm 0.05 \text{ mg cm}^{-1}$ ) was estimated by weighing the thermoplastic wheat gluten film before and after recollection of the electrospun coatings.

### 2.3. Characterization of films

#### 2.3.1. Scanning electron microscopy (SEM)

SEM was conducted on a Hitachi microscope (Hitachi S-4100) at an accelerating voltage of 10 kV and a working distance of 12–16 mm. Thermoplastic wheat gluten films as well as the

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