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Barrier properties and storage stability of edible coatings prepared with electrospraying

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ABSTRACT

Electrospraying is a novel technique for the application of coating to foods. In this study, thin lipid-based coatings were prepared by electrospraying on model surface and evaluated for their moisture barrier functionality. Sunflower oil and chocolate based coating materials were electrosprayed at elevated temperature (60 °C) using a multiple nozzle system. Sunflower oil coated the sides and top surface of the target surfaces, while chocolate based material deposited primarily on the top surface. In chocolate based coatings, larger droplet size and related lower charge to mass ratio explained the limited "wrap-around effect". Sunflower oil based coating penetrated into the target surfaces, which could be reduced by the addition of stearic acid (up to 0.15 g/g). However, this addition resulted in crystallization and crack formation during storage, and ultimately reduced barrier functionality. Conversely, chocolate-based material produced thicker coatings (up to 0.3 mm), which were more stable during storage and exhibited enhanced barrier properties.

Industrial relevance: Electrospraying is an efficient coating technique which can reduce the processing costs for industrial processes. This technique has been successfully applied to increase the shelf life of a minimally processed food. The results found in this study can be used at industry to obtain food products with desired sensory attributes along with prolonged shelf life.

1. Introduction

Electrospraying is an effective technique to atomize liquids, in which an electric potential difference is applied across a droplet emerging from a capillary. The electrical field induces accumulation of charge near the surface of the nascent droplet, and destabilizes the surface of droplet which disrupts into multiple diminutive charged droplets (Jaworek, 2007, 2008). These newly formed droplets do not coalesce in the air because of their charge and move along the electric field lines to the nearest grounded target surface. Electrospraying exhibits a higher transfer efficiency (80–90%) compared to conventional spraying methods which produce uncharged droplets (Jaworek & Sobczyk, 2008; Luo, Loh, Stride, & Edirisinghe, 2012; Oh, Kim, & Kim, 2008).

Ideally, in electrospraying, the charge of droplets leaks away upon contact with the target object. This is critical for smooth deposition of subsequent droplets (Khan, Mujawar, Schutyser, Schroën, & Boom, 2012; Khan, Schutyser, Schroën, & Boom, 2012b). The capability of electrospraying to produce micron-size charged droplets with a small size distribution makes the technique promising for coating purposes. The droplet size, deposition rate, and layer thickness can be controlled by optimizing process parameters such as flow rate and coating material properties (conductivity) (Jaworek & Sobczyk, 2008; Khan, Schutyser et al., 2012b).

Coating materials for foods can be formulated from lipids, proteins, polysaccharides or any combination of these. Lipids are excellent barriers against moisture migration (Debeaufort & Voilley, 2009; Khan, Mujawar et al., 2012). Their use as a protective layer dates back to the 12th century, when wax was applied to citrus fruits to increase their shelf-life (Debeaufort, Quezada-Gallo, & Voilley, 1998). Lipids, combined with proteins and polysaccharides, produce coatings with better mechanical and barrier properties. Besides, these composite films may have higher moisture permeability compared to that of pure lipid (Bravin, Peressini, & Sensidoni, 2004).

Lipid-based materials have been successfully electrosprayed (Gorty & Barringer, 2011; Khan, Mujawar, et al., 2012; Khan, Schutyser, et al., 2012b; Luo et al., 2012; Marthina & Barringer, 2012). However, to the best of our knowledge, electrosprayed coatings have not been investigated for their barrier functionality on food surfaces. Therefore, our study aims at evaluation of electrospraying as a method to prepare lipid-based coatings with vapor barrier properties on a model food target. For this, sunflower oil and chocolate-based coatings were electrosprayed on candy tablets (as model for other foods such as various bakery products) moving on a conveyer belt. Initially, the coating and spreading behaviors on the target surface were studied with respect

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Table 1
An overview of the different compositions that were used as coating material and corresponding values of their viscosity.

No	Lecithin (g/g)	Stearic acid (g/g)	PGPR (g/g)	Butter (g/g)	Sunflower oil (g/g)	Chocolate (g/g)	Viscosity (mPa·s)
1	0.20	0.10	–	–	0.70	–	20 ± 5
2	0.20	0.15	–	–	0.65	–	30 ± 6
3	0.20	–	0.15		–	0.65	420 ± 150
4	0.05	–	0.15	0.15	–	0.65	200 ± 7
5	0.05	–	0.15	0.30	–	0.50	140 ± 7

to the coating material properties. Latter, water vapor permeability of the coated tablets was investigated and compared to conventional 'dip'-coating.

2. Materials and methods

2.1. Film preparation

Sunflower oil and dark chocolate (Verkade Zaandam, Holland) were obtained from a local supermarket. Due to the low melting point of sunflower oil, stearic acid (Sigma-Aldrich Co.) was added to ensure solidification at room temperature. Alcolic-S Lecithin (American lecithin company, Oxford, CT, USA) was used as an additive to increase the conductivity of coating materials in order to facilitate electrospraying. Polyglycerol polyricinoleate (PGPR) (Givaudan, Vernier, Switzerland) and butter (SOP int. Ltd., UK) were added to the chocolate to reduce the viscosity. A summary of the compositions of sunflower oil and chocolate-based coatings is presented in Table 1. The viscosity of the coating material was measured with a rheometer (MCR 301, Anton Paar, Graz, Austria) with DG 26.7 geometry. A shear rate sweep was applied from 1 to 3000 s^{-1} and 3000 to 1 s^{-1} at a controlled temperature of 60 °C.

Readily available hygroscopic candy tablets (brand name 'zwartwit', Fortuin Dokkum, The Netherlands) were used to evaluate the coatings applied by both electrospraying and dipping (as reference). The initial water activity (a_w) of a packaged tablet is approximately 0.40 and it thus absorbs water when exposed to relative humidity higher than 40%. The tablets were coated by electrospraying on a conveyer belt at a velocity of 1 mm/s; the mass of applied coating material was measured after various (1–6) passes.

A multi-nozzle electrospraying system (Terronics Development Cooperation, USA) was used to prepare films at 60 °C ± 5 °C in a temperature-controlled cabinet. The nozzles were subjected to an electric potential of 20–25 kV using a high voltage source (Heinzinger electronic GmbH). The coating material was supplied at a rate of 0.9 ml/min via a syringe pump (Harvard 11 plus, Harvard Inc., USA). As a reference, a set of three tablets were dipped in the coating material; the applied amounts ranged from 0.1 to 0.9 kg/m^2. The coated tablets were subsequently cooled down at ambient temperature to solidify the coatings.

2.2. Film analysis

A set of three coated and uncoated tablets were analyzed for moisture uptake as a function of time at 60% relative humidity and 20 °C in a humidity chamber (Memmert GmbH). The water activity difference caused the moisture uptake which was monitored daily by weighing the tablets. A minimum of three replicates were measured for each film preparation and the average results of relative water uptake were reported. The relative flux reduction was calculated as described in Eq. (1):

$$\text{Relative flux reduction} = 1 - \frac{M_c}{M_b} \tag{1}$$

where M_c and M_b are the mass increase as function of time (g/h) of the coated and uncoated tablets, respectively. Additionally, Water Vapor Permeability (WVP) and Permeance (P) of the coatings were calculated by Eqs. (2) & (3):

$$\text{WVP} = \frac{\Delta M\, L}{A \cdot \Delta p_v} \tag{2}$$

$$P = \frac{\Delta M}{A \cdot \Delta p_v} \tag{3}$$

where ΔM is the mass increase as function of time (g/s) of the coated tablets, L is the film thickness, A is the coated area available for water vapor migration (m^2), and Δp_v is the vapor pressure difference across the film (Pa) (Boom, Schroen, & Vermue, 2011). All measurements were performed in triplicate and values were reported as mean with standard deviation. During the storage, the crack and pinhole formation in coatings was studied with a light microscope (Axiovert 200 MAT, Carl Zeiss B.V., Sliedrecht, The Netherlands) attached to a camera (MotionPro HS4, Redlake MASD Inc., San Diego, CA, USA).

To measure the penetration of coatings inside the tablets, Sudan red was used to label the sunflower oil-based coatings, which were analyzed with a confocal scanning laser microscope (CSLM) (Carl Zeiss Axiovert 200 microscope, Zeiss, Jena, Germany) equipped with a LSM 5 Exciter and He–Ne laser lamp operating at a wavelength of 633 nm.

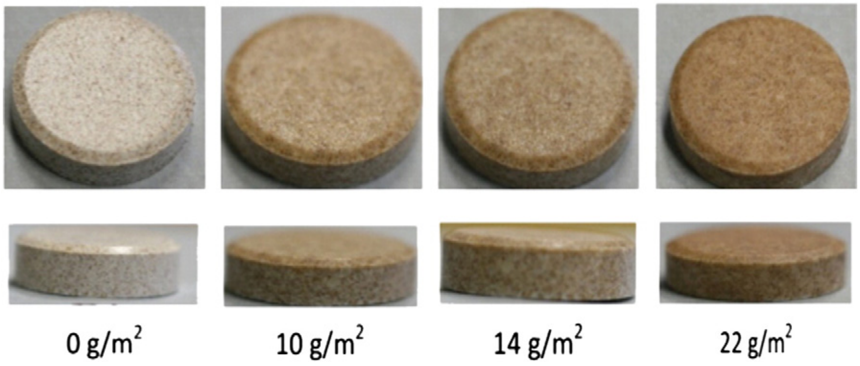

Fig. 1. Visual representation of 'zwartwit' tablets coated with increasing amounts of sunflower oil based coating applied by electrospraying.