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# Wettability of gelatin coating formulations containing cellulose nanofibers on banana and eggplant epicarps

R. Andrade [a,b,*], O. Skurtys [c], F. Osorio [a], R. Zuluaga [d], P. Gañán [e], C. Castro [f]

[a] Department of Food Science and Technology, Universidad de Santiago de Chile, Avenida Ecuador 3769, Santiago, Chile
[b] Department Food Engineering, Universidad de Cordoba, Carrera 6 N° 76-103, Montería, Colombia
[c] Department of Mechanical Engineering, Universidad Técnica Federico Santa María, Av. Vicuña Mackenna 3939, Santiago, Chile
[d] Facultad de Ingeniería Agroindustrial, Universidad Pontificia Bolivariana, Circular 1° N° 70-01, Medellín, Colombia
[e] Facultad de Ingeniería Mecánica, Universidad Pontificia Bolivariana, Circular 1° N° 70-01, Medellín, Colombia
[f] Facultad de Ingeniería Textil, Universidad Pontificia Bolivariana, Circular 1° N° 70-01, Medellín, Colombia

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## ABSTRACT

An important physical parameter used to characterize the adhesive properties of the coating formulation is the surface free energy (SFE), which is a key parameter of the wetting capacity of a food surface. The objective of this work was to determine the effects of the concentrations of gelatin (0.6–2 g/100 mL), glycerol (10–20 g/100 g) and nanofiber cellulose (1–5 g/100 g) on the wettability of gelatin-based edible coatings on banana and eggplant epicarps, and apply the response surface method to optimize the coating formulation. The SFE of banana and eggplant epicarp was calculated by Zisman plot and acid-base methods. Spreading coefficients of the coating formulations were determined on both epicarps. Banana epicarp was more hydrophilic than eggplant epicarp, but both surfaces are a low-energy surfaces and slightly bipolar. The cohesive energy of the coating formulations was influenced significantly by gelatin and cellulose nanofiber concentrations. For both epicarps, addition of glycerol and cellulose nanofibers enhanced the wetting of coating formulations based on gelatin. The best formulations in which the spreading coefficient of coating formulations on banana and eggplant epicarps reached a maximum of $-22.44$ mN m$^{-1}$ and $-32.95$ mN m$^{-1}$, respectively.

## 1. Introduction

Edible coatings help meet many challenges related to the storage and marketing of food products. The functionality and performance of edible coatings depend on their barrier and mechanical properties, which in turn depend on film composition, its formation process and the method of application on the food (Andrade, Skurtys, & Osorio, 2012; Lin & Zhao, 2007).

Edible coatings are made from biological materials like proteins (gelatin, casein, zein), polysaccharides (starch, cellulose, alginate), and lipids (beeswax, fatty acids). Gelatin is well known, among the animal proteins, for its film forming properties (Krochta, 2002 , 1–32). Gelatin coatings show good barrier characteristics against oxygen and aroma transfers at low and intermediate relative humidity. However, they have poor barrier properties against water vapor transfer due to their hydrophilic nature (Carvalho et al., 2008; Jongjareonrak, Benjakul, Visessanguan, Prodpran, & Tanaka, 2006; Limpisophon, Tanaka, Weng, Abe, & Osako, 2009). Many attempts have been made to modify the poor properties of protein films, including compounding with natural fibers dispersed in the biopolymer matrix. For example, George and Siddaramaiah (2012) reported that addition of bacterial cellulose nanocrystals reduced moisture transport in gelatin-based films.

The coating process involves wetting of the food to be coated by the coating formulation, possible penetration of the solution into the peel, followed by a possible adhesion between these two materials (Hershko, Klein, & Nussinovith, 1996). The coating must be designed considering food surface properties, especially surface energy, in order to satisfy adequate adherence and thickness on the food surface. With an adequate spread of the coating solution on the food surface, it is possible to avoid interspaces between the food and the coating, preventing thicker film zones that can produce anaerobic conditions that would lead to food deterioration. The affinity between the food surface and coating formulation is

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* Corresponding author. Department of Food Science and Technology, Universidad de Santiago de Chile, Avenida Ecuador 3769, Santiago, Chile. Tel.: +56 27180600.

*E-mail addresses:* ricardo.andrade@usach.cl, rdandrade@correo.unicordoba.edu.co (R. Andrade).

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fundamental in the coating design, considering that the effective spreading of a coating on a food is greatly influenced by the wettability of the surface by the coating formulation (Andrade, Skurtys, & Osorio, 2013; Choi, Park, Ahn, Lee, & Lee, 2002; da Cunha et al., 2009; Hong, Han, & Krochta, 2004; Ribeiro, Vicente, Teixeira, & Miranda, 2007).

Wettability can be defined as the tendency for a coating formulation to spread on a food surface. It can be characterized by the degree and the rate of wetting. The rate of wetting indicates how fast the liquid spreads on the surface, while the degree of wetting indicates the extent to which the coating formulation wets the food surface. The main parameter used to characterize the wetting of a food surface is the equilibrium contact angle ($\theta$) presented in Fig. 1 (Lee, Ivanova, Starov, Hilal, & Dutschk, 2008; Miller & Neogi, 2008). In general, when a liquid coating formulation is placed on a food surface, the coating may spread over the food surface, i.e. it completely wets it (Fig. 1a), or it forms a finite contact angle with the food surface. When the contact angle is between 0 and 90° the situation is referred to as partial wetting (Fig. 1b), whereas if the contact angle is greater than 90° the liquid does not wet the surface, a situation refereed to as non-wetting (Fig. 1c). The distinction between the different wetting states is usually made by considering the equilibrium spreading coefficient ($S_{\text{cf/food}}$) determined from the balance between the adhesive forces ($W_A$) of the coating formulation on the food surface and the cohesive forces ($W_C$) of the coating formulation (Eq. (1)).

$$S_{\text{cf/food}} = W_A - W_C \tag{1}$$

where $S_{\text{cf/food}}$ is the spreading coefficient of the coating formulation on a food surface, $W_A$ and $W_C$ are the adhesion and cohesion energies, defined by Eqs. (2) and (3), respectively:

$$W_A = \gamma_{SV} + \gamma_{LV} - \gamma_{SL} = \gamma_{LV}(1 + cos\theta) \tag{2}$$

$$W_C = 2\gamma_{LV} \tag{3}$$

where $\gamma_{SV}$ is the interfacial tension between the food surface and the air, $\gamma_{LV}$ is the interfacial tension between the liquid coating formulation and the air, $\gamma_{SL}$ is the interfacial tension between the food surface and the liquid coating solution, and $\theta$ is the equilibrium contact angle. Each of these forces acts in different form. Adhesive forces cause the liquid to spread on the food surface, while the cohesive forces allow the coating formulation and the food surface to remain together (Lee et al., 2008; Miller & Neogi, 2008).

An important physical parameter used to characterize and predict adhesive properties of the coating formulation is the surface free energy (SFE), which is a key parameter of the wetting capacity of the food surface. Food surfaces with low SFE ($<$75 mN m$^{-1}$) are called "low energy" or hydrophilic surfaces (Koopal, 2012). For a low energy of the food surface, depending on the coating formulation used, it is possible to obtain partial or complete wetting.

The main methods for the determination of SFE are those of Zisman and van Oss-Chaudhury–Good (vOCG). Zisman's method uses an estimation of the critical surface tension ($\gamma_C$) of the food surface, by extrapolation from the Zisman plot. For a simple molecular liquid, $\gamma_C$ of the food surface is essentially independent of the nature of the liquid and is characteristic of the food surface alone (Casariego et al., 2008; de Gennes, 1985; Zisman, 1964). In food engineering, Zisman's method has been used for a long time. For different foods, it is possible to find various $\gamma_C$ values in the literature (Cerqueira, Lima, Teixeira, Moreira, & Vicente, 2009; Choi et al., 2002; Ribeiro et al., 2007; Skurtys et al., 2011). The vOCG method, also known as the acid-base approach (Eq. (4)), gives more details on the food surface free energy than the Zisman method. The vOGC method separates the SFE into two components, long-range Lifshitz–van der Waals (LW) interactions and short-range polar interactions (electron acceptor, $\gamma_{SV}^{+}$, and electron donor, $\gamma_{SV}^{-}$) (Keijbets, Chen, Dickinson, & Vieira, 2009; Zenkiewicz, 2007).

$$\gamma_{LV}(1 + \cos\theta) = 2\left(\sqrt{\gamma_{SV}^{LW}\gamma_{LV}^{LW}} + \sqrt{\gamma_{SV}^{+}\gamma_{LV}^{-}} + \sqrt{\gamma_{SV}^{-}\gamma_{LV}^{+}}\right) \tag{4}$$

To obtain the $\gamma_{SV}^{LW}$, $\gamma_{SV}^{+}$, $\gamma_{SV}^{-}$ parameters of the food surface, a system of three equations (Eq. (4)) are solved simultaneously after measuring the contact angle data of three different liquids, two of them being polar.

The present work was aimed at studying the influence of gelatin, glycerol, and bacterial cellulose concentrations on the wettability of two fruits epicarp that have different surface free energy: banana and eggplant.

## 2. Materials and methods

### 2.1. Plant materials

Bananas and eggplants were purchased as fresh fruits from the local market (Santiago, Chile). The fruits were carefully selected for their uniformity in ripeness, size, color, and absence of physical damage on their surface according to visual analysis. Before measurements, the fruits were left at room temperature (20 ± 2 °C) and rectangular samples were cut: 1.5 cm × 2.5 cm.

### 2.2. Materials

Type B gelatin from bovine skin (180 Bloom) was purchased from Rousselot (Rousselot, Brazil), glycerol was purchased from Sigma (Sigma–Aldrich, Chile). Cellulose nanofibers were obtained from agroindustrial residues (pineapple peel juice), and *Gluconacetobacter swingsii* sp. as reported by Castro et al. (2011). The culture medium used for bacterial cellulose production was pineapple peel juice (2.14 g/100 mL glucose, 2.4 g/100 mL fructose, 2.10 g/

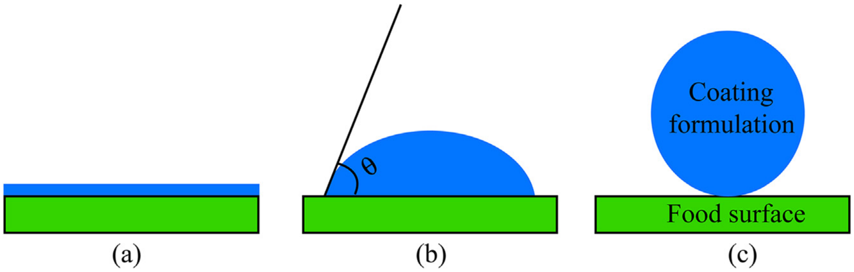

**Fig. 1.** Liquid coating formulation on a food surface. (a) Total wetting, hydrophilic food ($\theta = 0°$); (b) Partial wetting ($0 < \theta < 90°$); (c) No wetting, hydrophobic food ($\theta > 90°$).