

# Comparison of the deep frying process in coated and uncoated dough systems

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## Abstract

The effect of an edible methylcellulose coating to reduce oil uptake during frying was analyzed on a dough system. The oil uptake reduction was 30% for coated dough discs compared to uncoated ones; the coating neither modified the water content of the samples nor the quality attributes of the fried dough such as color and texture.

A mathematical model was developed including simultaneous heat and water vapor transfer at the different stages of the frying process and the cooling at environmental conditions after the product was removed from the fryer. It allowed to simulate satisfactorily the experimental data of temperature profiles, water losses and the thickness of the dehydrated zone ( $d_T$ ) as a function of frying time. Post-frying oil uptake was correlated with  $d_T$  considering microstructural changes during frying time, analyzed by SEM (scanning electron microscopy). The coating reduced oil uptake, modifying the wetting properties related to the interfacial tension and also becoming a mechanical barrier to lipids.

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## 1. Introduction

The reduction of the lipid content in fried foods is required mainly owing to its relation with obesity and coronary diseases. An alternative to reduce oil uptake in fried foods is the use of edible films or coatings. The application of hydrocolloid coatings allows to reduce oil content of deep-fat fried products due to its lipid barrier properties; the most widely studied are gellan and cellulose derivatives (Mellema, 2003; Williams & Mittal, 1999a, 1999b). Cellulose derivatives, including methylcellulose (MC) and hydroxypropyl-methylcellulose (HPMC) exhibit thermogelation; when suspensions are heated they form a gel that reverts below the gelation temperature, and the original

suspension viscosity is recovered. Mallikarjunan, Chinnan, Balasubramaniam, and Phillips (1997) and Huse, Mallikarjunan, Chinnan, Hung, and Phillips (1998) demonstrated the effectiveness of various edible coatings in reducing oil absorption in starchy products.

Frying phenomena occur during the immersion of the product in oil at a temperature of 150–200 °C, where a simultaneous heat and mass transfer take place (Singh, 1995; Aguilera & Hernández, 2000). The mechanism of oil penetration is a subject of controversy (Pinthus & Saguy, 1994; Ufheil & Escher, 1996; Kassama, 2003; Mellema, 2003). Many researchers have suggested that oil absorption on the surface of the fried product occurs when samples are removed from the frying medium (Bouchon, Aguilera, & Pyle, 2003; Bouchon & Pyle, 2005a, 2005b; Moreira & Barrufet, 1998; Perkins & Erikson, 1996; Williams & Mittal, 1999a, 1999b; Yamsaengsung & Moreira, 2002a, 2002b). Conditions at which products are removed from the frying oil seem decisive for the uptake of oil; this

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## Nomenclature

|           |   |                      |   |
|-----------|---|----------------------|---|
| $A$       | area (m <sup>2</sup> )  | WC                   | water content of the samples in dry basis (g water/g dry solid) |
| $c_p$     | specific heat (J/(kg °C))   | $x$                  | spatial coordinate (m)  |
| $C_v$     | concentration of water vapor in the dehydrated zone (kg/m <sup>3</sup> )            | $x_1$                | position of the vaporization front (m)                          |
| $C_w$     | concentration of water in the dough (kg/m <sup>3</sup> )                            | <i>Greek symbols</i> |   |
| $d$       | distance from surface (m)   | $\varepsilon$        | porosity  |
| $D_v$     | effective diffusion coefficient of vapor in the dehydrated food (m <sup>2</sup> /s) | $\tau$               | tortuosity factor   |
| $D_w$     | effective diffusion coefficient of water in the food matrix (m <sup>2</sup> /s)     | $\alpha$             | thermal diffusivity (m <sup>2</sup> /s)                         |
| $d_T$     | thickness of the dehydrated zone (m)  | $\rho$               | density (kg/m <sup>3</sup> )                                    |
| $h$       | heat transfer coefficient (W/(m <sup>2</sup> K))                                    | <i>Subscripts</i>    |   |
| $k$       | thermal conductivity (W/(m K))  | a                    | air   |
| $L$       | half thickness of the dough sample (m)  | d                    | dehydrated  |
| $L_{vap}$ | latent heat of vaporization of water (J/kg)   | ave                  | average   |
| $L^*$     | lightness   | ini                  | initial   |
| OU        | oil uptake in dry basis (g oil/g dry solid)   | w                    | water   |
| OUR       | oil uptake relative variation (%)   | o                    | oil   |
| $t$       | time, s (or min)  | v                    | vapor   |
| $T$       | temperature (°C)  | va                   | vapor–air   |
| $V$       | volume of the sample (cm <sup>3</sup> )   | vap                  | vaporization  |
| WR        | water retention relative variation (%)  |                      |   |

would be related to the adhesion of oil to the surface and draining phenomena.

Several models have been developed for heat, moisture and fat transfer during frying of foods (Ni & Datta, 1999; Yamsaengsung & Moreira, 2002a, 2002b). Ateba and Mittal (1994) developed a model for heat, moisture and fat transfer in deep-fat frying of beef meatballs. Farakas, Singh, and Rumsey (1996a, 1996b) developed a model for frying considering heat and moisture transfer and Singh (2000) treated the crust as a moving boundary.

The objectives of the present work were:

1. To analyze the performance of applying an edible coating based on methylcellulose (MC) on a food model dough system by measuring moisture content and oil uptake in deep-frying process.
2. To compare the quality attributes of the fried coated and uncoated products evaluating texture and color.
3. To observe by Scanning Electron Microscopy the surface microstructure of coated and uncoated fried products at different processing times to assess the effect of the MC coating.
4. To model heat and moisture transfer during the deep-fat frying process of food systems (with and without MC coating), solving numerically the partial differential equations that allow to predict temperature profiles and water concentrations, as a function of operating conditions.
5. To validate experimentally the mathematical model evaluating the temperature profiles and the water losses.

6. To relate oil uptake measurements after the frying process with the effect of frying time on sample microstructural changes and the growth of the dehydrated zone.

## 2. Materials and methods

### 2.1. Model dough system

The food model system used was prepared with 200 g refined wheat flour (Molinos Río de La Plata, Argentina) and 110 ml distilled water, approximately. From the obtained dough, discs of 60 mm in diameter and 7 mm thick were cut and immediately used. For each experiment, 6 control and 6 coated discs were fried.

### 2.2. Edible coating formulation

Methylcellulose, MC (A4M, Methocel) was provided by COLORCON S.A. (Argentina). A 1% (w/w) MC aqueous solution was used and 0.75% (w/w) sorbitol (Merck, USA) was added as plasticizer. MC and sorbitol concentrations were selected in a previous work (García, Ferrero, Bértola, Martino, & Zaritzky, 2002).

### 2.3. Coating procedure and frying experiments

Dough samples were dipped in the coating suspensions for 30 s and immediately fried. Uncoated (control) and coated samples were fried in a controlled temperature

deep-fat fryer (Yelmo, Argentina) filled with 1.5 L of commercial sunflower oil (AGD, Córdoba, Argentina). Oil composition was 99.93% lipids, with 25.71% mono-unsaturated and 64.29% poly-unsaturated fatty acids. Used oil was replaced by fresh oil after four frying batches; in each batch up to four dough discs were fried, maintaining in all experiments the same product–oil volume ratio.

Different constant frying temperatures were tested to select the working frying conditions according to sample characteristics. These temperatures ranged between  $150 \pm 0.5$  °C and  $170 \pm 0.5$  °C; frying times ranged between 5 and 15 min. Optimum time–temperature frying conditions were determined by a non-trained sensory panel of 6 members; panelists judged color, flavor, texture and overall appearance of the samples as described in a previous work (García et al., 2002).

#### 2.4. Quality attributes

To evaluate whether MC coating application affected the quality attributes, color and texture parameters of coated and uncoated fried dough samples, were analyzed.

##### 2.4.1. Colorimetric measurements

Assays were carried out with a Minolta colorimeter CR 300 Series (Japan). The CIELab scale was used, lightness ( $L^*$ ) and chromaticity parameters  $a^*$  and  $b^*$  were measured.  $L^*$ ,  $C^*$  (chroma),  $H^\circ$  (Hue) and color differences ( $\Delta E$ ) were also calculated as

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (1)$$

where

$$\Delta L^* = L_0^* - L_t^*, \quad \Delta a^* = a_0^* - a_t^*, \quad \Delta b^* = b_0^* - b_t^*$$

being  $L_t^*$ ,  $a_t^*$ ,  $b_t^*$ , the color parameter values of samples fried at different frying times.  $L_0^*$ ,  $a_0^*$ ,  $b_0^*$  were selected as the color parameters of samples fried for 3 min at 160 °C and not the color of the raw sample in order to analyze the effect of frying time.

Samples were analyzed in triplicates, recording four measurements for each sample.

##### 2.4.2. Texture analysis

Breaking force of samples was measured by puncture test using a texture analyzer TA.XT2i – *Stable Micro Systems* (Haslemere, Surrey, UK) with a 5 kg cell. Samples were punctured with a cylindrical plunger (2 mm diameter) at 0.5 mm/s. Maximum force at rupture was determined from the force–deformation curves. At least 10 samples were measured for each assay. Samples were allowed to reach room temperature before performing the tests.

#### 2.5. Water content

Water content (WC) was determined measuring weight loss of fried products, upon drying in an oven at 110 °C

until constant weight (García et al., 2002). At different frying times the relative variation of water retention % (WR) in the coated product relative to the uncoated one was calculated as follows:

$$WR = \left( \frac{WC \text{ coated}}{WC \text{ uncoated}} - 1 \right) \times 100 \quad (2)$$

For each frying time condition, results were obtained using all the samples from at least two different batches. The equilibrium water content was defined as the humidity reached at long frying times (1080 s).

#### 2.6. Oil uptake

Oil uptake (OU) of fried products was determined measuring the lipid content of dried samples using a combined technique of successive batch and semi-continuous Soxhlet extractions. The first batch extraction was performed with petroleum ether:ethyl ether (1:1) followed by a Soxhlet extraction with the same mixture and another with *n*-hexane. Oil uptake relative variation % (OUR) in the coated product relative to the uncoated one was calculated as follows:

$$OUR = \left( 1 - \frac{OU \text{ coated}}{OU \text{ uncoated}} \right) \times 100 \quad (3)$$

For each frying time condition, results were obtained using all the samples from at least two different batches.

#### 2.7. Microscopy observations

Scanning electron microscopy (SEM) of coated and uncoated samples was performed with a JEOL JSMP 100 scanning electron microscope (Japan). Coated pieces were mounted on bronze stubs using double-sided tape and then coated with a layer of gold (40–50 nm). All samples were examined using an accelerating voltage of 5 kV.

The microstructure of the fried samples was also analyzed using an environmental electronic microscope Electroscan ESEM 2010 (ESEM, Environmental Scanning Electron Microscopy). The effects of frying time and coating application on the structure were studied.

#### 2.8. Thermal histories of samples during frying process

Sample temperatures of coated and uncoated samples as a function of frying time were measured using copper–constantan thermocouples (Omega). Temperature measurements in the center of the dough were performed by introducing the thermocouple carefully through the disc from the border towards the center; besides, the temperature of an internal sample point and the temperature of the frying medium were also recorded.

Temperatures were measured and recorded each 5 s using type T thermocouples linked to a data acquisition and control system Keithley KDAC Series 500 (Orlando FL, USA).

## 2.9. Mathematical model of heat and moisture transfer during frying process of a dough system

Deep-fat frying is a complex process that involves simultaneous heat and mass transfer. The process induces a variety of physicochemical changes in both the food and the frying medium.

A mathematical model of the frying process was proposed based on the numerical solution of heat and mass transfer partial differential equations under unsteady state conditions. The frying process was modeled in dough discs with and without coating. Discs were considered as infinite slabs and unidirectional, heat and mass transfer equations were solved for  $0 \leq x \leq L$ . In the proposed model, different stages were considered.

*Stage 1:* represents the initial heating of the product in which energy and mass transfer between the product and the frying oil occurs. The model considered an initial uniform temperature distribution in the sample ( $T_{ini}$ ).

To get the temperature profiles the microscopic energy balance was solved:

$$\rho c_p \frac{\partial T}{\partial t} = \nabla(k \nabla T) \quad (4)$$

Thermal properties corresponded initially to the raw dough and changed with temperature and moisture content (Table 1).

The initial and boundary conditions were:

$$t = 0 \quad T = T_{ini} \quad 0 \leq x \leq L, \quad (5)$$

$$x = 0 \quad \frac{\partial T}{\partial x} = 0 \quad t > 0, \quad (6)$$

$$x = L \quad -k \frac{\partial T}{\partial x} = h_1(T - T_{oil}) + L_{vap} m_{vap,1} \quad t > 0 \quad (7)$$

where  $m_{vap,1}$  is the water flux from the product and  $h_1$  is the convective heat transfer coefficient. The heat transfer

mechanism was considered governed by natural convection ( $h_1 = 50 \text{ W/m}^2\text{K}$ ).

In order to evaluate water profiles the microscopic mass balance was solved:

$$\frac{\partial C_w}{\partial t} = \nabla(D_w \nabla C_w) \quad (8)$$

The following initial and boundary conditions were considered:

$$t = 0 \quad C_w = C_{w,ini} \quad 0 \leq x \leq L \quad (9)$$

$$x = 0 \quad \frac{\partial C_w}{\partial x} = 0 \quad t > 0 \quad (10)$$

$$x = L \quad -D_w \frac{\partial C_w}{\partial x} = m_{vap,1} \quad t > 0 \quad (11)$$

$m_{vap,1}$  is calculated from Eq. (11) with  $C_w = 0$  at the sample/oil interface, because it was considered that water evaporates and immediately leaves the frying medium.

For each time interval the model calculated the average values of temperature and moisture content as follows:

$$T_{ave} = \frac{\int_0^L T dV}{V} \quad (12)$$

$$C_{w,ave} = \frac{\int_0^L C_w dV}{V} \quad (13)$$

The second stage starts when the surface temperature of the product reaches that of water vaporization.

*Stage 2:* Water boiling is produced on the surface and a dehydrated layer is formed in the external zone of the product, having different thermo-physical and transport properties from those of the wet core (Table 1). Several authors (Bouchon et al., 2003; Bouchon & Pyle, 2005a, 2005b; Farkas et al., 1996a, 1996b; Ni & Datta, 1999) used the denomination of crust for the dehydrated zone. In the present work we considered that crust is only the portion of the dehydrated zone containing oil.

When water on the surface is no longer available, vaporization occurs in a front that moves towards the inner zone of the product. During this phase of the frying process, the vapor being released from the product surface impedes oil penetration into the product (Aguilera & Hernández, 2000; Kassama, 2003; Singh, 1995).

Microscopic energy balances given by Eq. (4) were solved for both (core and dehydrated) zones to obtain temperature profiles, considering the corresponding thermal properties (Table 1).

The initial temperature profile in this stage was given by the final profile obtained at the end of Stage 1. The following boundary conditions were applied:

$$x = 0 \quad \frac{\partial T}{\partial x} = 0 \quad t > 0 \quad (14)$$

$$x = x_1 \quad T = T_{vap} \quad t > 0 \quad (15)$$

$$x = L \quad -k_d \frac{\partial T}{\partial x} = h_2(T - T_{oil}) \quad t > 0 \quad (16)$$

With  $x_1$  = the position of the vaporization front (measured from the center of the slab).

Table 1  
Core, dehydrated zone, and oil properties used in the model

| Property  | Core zone   | Dehydrated zone   | Sunflower oil     |
|---|---|-------------------|-------------------|
| Initial water content   | 0.42  | –                 | –                 |
| Density (kg/m <sup>3</sup> )  | 623 <sup>d</sup>  | 579 <sup>a</sup>  | 876 <sup>c</sup>  |
| Thermal conductivity (W/(m K))  | 0.6 <sup>d</sup>  | 0.05 <sup>a</sup> | 0.6 <sup>c</sup>  |
| Specific heat (J/kg °C)   | 2800 <sup>d</sup>   | 2310 <sup>a</sup> | 2033 <sup>c</sup> |
| Porosity  | –   | 0.42 <sup>b</sup> | –                 |
| Tortuosity  | –   | 8.8 <sup>b</sup>  | –                 |
| Diffusion coefficient of vapor in air (dehydrated zone) (m <sup>2</sup> /s) | $\frac{5.6810^{-9}(T + 273.16)^{1.5}}{1.789 - 2.1310^{-3}(T + 273.16)} e$ |                   |                   |
| Moisture diffusion coefficient in dough (core zone) (m <sup>2</sup> /s)     | $2 \times 10^{-9f}$   |                   |                   |

<sup>a</sup> Rask (1989).

<sup>b</sup> this work.

<sup>c</sup> Moreira et al. (1999).

<sup>d</sup> Zanoni et al. (1995).

<sup>e</sup> Bird et al. (1976).

<sup>f</sup> Tong and Lund (1990).



Besides the thickness of the dehydrated zone ( $d_T$ ), measured from the sample/oil interface can be calculated as  $d_T = L - x_1$ .

Values of the heat transfer coefficient ( $h_2$ ) higher than  $250 \text{ W/m}^2 \text{ K}$  were found in literature for the whole frying process (Costa, Oliveira, Delaney, & Gekas, 1998; Dager-skog & Sorenfor, 1978; Moreira, Palau, Sweat, & Sun, 1995). These high heat transfer coefficients allowed us to assume a constant temperature at the product oil interface.

Moisture content decreased due to water vaporization; vapor diffused through the dehydrated zone which was considered as a porous medium.

The following mass balance was solved in the dehydrated zone:

$$m_{\text{vap},2} = -D_v \frac{\partial C_v}{\partial x} \quad (17)$$

$D_v$  was calculated as follows:

$$D_v = D_{\text{va}} \frac{\varepsilon}{\tau} \quad (18)$$

The following conditions were considered:

$$x = x_1 \quad C_v = C_{v,\text{equi}} \quad (19)$$

$$x = L \quad C_v = 0 \quad (20)$$

where  $C_{v,\text{equi}}$  is the equilibrium vapor concentration at  $T_{\text{vap}}$ ; it was calculated using Clausius–Clapeyron equation assuming ideal gas behavior for the water vapor.

To calculate the position of the vaporization front as a function of time, the following equation was proposed:

$$-C_{w,\text{ave}} \frac{dx_1}{dt} = -D_v \frac{\partial C_v}{\partial x} \quad (21)$$

$$\text{then } \frac{dx_1}{dt} = \frac{D_v}{C_{w,\text{ave}}} \frac{\partial C_v}{\partial x} \quad (22)$$

When the vaporization front reaches the center of the fried product, the dehydrated food system increases its temperature until the process is completed. Water transfer is absent and the moisture content corresponds to the bound water. To estimate temperature profiles the microscopic energy balance (Eq. (4)) with the boundary conditions (6) and (16) was solved.

*Stage 3:* The final step corresponds to the cooling of the fried product when it is removed from the hot oil medium. The energy balance (Eq. (4)) was solved with the boundary condition (Eq. (6)) and considering a natural convection heat transfer coefficient ( $h_3 = 10 \text{ W/m}^2 \text{ K}$ ) at the food–air interface, as follows:

$$x = L \quad -k_d \frac{\partial T}{\partial x} = h_3(T - T_a) \quad t > 0 \quad (23)$$

In this step the oil uptake is produced.

Previous works (García et al., 2002; García, Ferrero, Bértola, Martino, & Zaritzky, 2004) showed that the average coating thickness determined by SEM was approximately  $10 \mu\text{m}$ . Then, the same heat transfer equations were applied for coated and uncoated products. Besides,

since methylcellulose coating is a hydrophilic polymer, poor water vapor barrier properties were expected. Considering this assumption, water transfer was described using the same equations as for uncoated systems.

The mathematical model was solved for the different stages. The microscopic energy and mass balances led to a system of coupled non-linear partial differential equations. The system was solved by an Implicit Finite-Differences Method (Crank–Nicolson centered method).

In the Stage 2 a variable grid was applied to overcome the deformation of the original fixed grid due to the shift of the vaporization front. The scheme used in the present work is similar to that developed for freezing and frozen storage (Campañone, Salvadori, & Mascheroni, 2001). The complete solution was codified in a Fortran 90 program. The model employed 20 nodes and an interval of time of 0.1 s.

### 2.10. Statistical analysis

Systat-software (SYSTAT, Inc., Evanston, IL, USA, 2001) version 10.0 was used for all statistical analysis. Analysis of variance (ANOVA), non-linear regression analysis and Fisher LSD mean comparison test were applied. The significance levels used were 0.05 and 0.01.

## 3. Results and discussion

### 3.1. Selection of the frying conditions

Frying conditions determine sensory characteristics and consumer acceptability of fried products, thus, sensory characterization was performed to select time–temperature conditions. In all cases, panelists could not distinguish between the coated and the uncoated samples. Sensory analysis (color, flavor, texture and overall appearance) determined that  $12 \text{ min}$  at  $160 \pm 0.5 \text{ }^\circ\text{C}$  were the best frying conditions for dough discs. Besides this was the time required to complete starch gelatinization in dough discs (confirmed by microscopy observation) and consequently fried samples could be considered cooked.

### 3.2. Comparison of the experimental moisture content and oil uptake of coated and uncoated fried products

MC coatings reduced the lipid content of dough discs significantly ( $P < 0.05$ ); the oil uptake was 30% lower than that corresponding to uncoated samples (Table 2). Similarly, Williams and Mittal (1999a) reported that MC coatings reduced the lipid content of potato spheres in 34.5% compared to the control samples. This result was attributed to the presence of hydrocolloid films acting as lipid barriers, particularly MC due to its thermal gelation properties. The oil uptake of coated and uncoated dough discs at each frying time are shown in Table 2. The final lipid content values, at long frying times, were  $0.0894 \text{ g oil/g dry solid}$  for uncoated samples and  $0.0626 \text{ g oil/g dry solid}$  for

Table 2  
Lipid and water contents of uncoated and coated dough discs as a function of frying time

| Time (s) | OU of uncoated samples (g oil/g dry solid) | OU of coated samples (g oil/g dry solid) | WC of uncoated samples (g water/g dry solid) | WC of coated samples (g water/g dry solid) | WR, water retention relative variation (%) | OUR (%) |
|----------|--|--|--|--|--|---------|
| 0        | 0.00                                       | 0.00                                     | 0.721 ± 0.043                                | 0.786 ± 0.059                              | 9.09                                       | –       |
| 180      | 0.053 ± 0.010                              | 0.045 ± 0.010                            | 0.492 ± 0.017                                | 0.477 ± 0.013                              | –3.06                                      | 14.07   |
| 360      | 0.058 ± 0.016                              | 0.049 ± 0.001                            | 0.356 ± 0.013                                | 0.352 ± 0.012                              | –1.05                                      | 15.36   |
| 540      | 0.073 ± 0.008                              | 0.054 ± 0.001                            | 0.300 ± 0.013                                | 0.296 ± 0.007                              | –1.34                                      | 26.36   |
| 720      | 0.074 ± 0.016                              | 0.053 ± 0.006                            | 0.253 ± 0.037                                | 0.200 ± 0.006                              | –21.12                                     | 29.07   |
| 900      | 0.082 ± 0.017                              | 0.062 ± 0.013                            | 0.142 ± 0.016                                | 0.159 ± 0.016                              | 11.43                                      | 24.62   |
| 1080     | 0.089 ± 0.005                              | 0.063 ± 0.005                            | 0.157 ± 0.005                                | 0.170 ± 0.004                              | 8.03                                       | 29.91   |

coated ones. Results showed that the ratio between the lipid content (dry basis) of uncoated and coated samples was 1.4 at 1080 s, verifying that the MC coating acted as an effective oil barrier.

With regard to water content, non-significant differences ( $P > 0.05$ ) were detected between coated and uncoated discs (Table 2); this could be attributed to the poor water vapor barrier of MC films (Donhowe & Fennema, 1993). The equilibrium water content values, which correspond to bound water, were 0.157 g water/g dry solid and 0.1695 g water/g dry solid for uncoated and coated samples, respectively.

### 3.3. Quality attributes of fried coated and uncoated samples

During the cooking process (for  $t < 720$  s) similar values of maximum force were obtained for coated and uncoated fried samples. In all cases, the maximum force to puncture the samples increased as a function of frying time, due to the formation of a dehydrated zone (Fig. 1a).

Non-significant ( $P > 0.05$ ) differences were observed in the analyzed color parameters of coated and uncoated dough discs. Chromaticity parameter  $b^*$  increased significantly ( $P < 0.05$ ) with frying time, while lightness ( $L^*$ ) and chromaticity parameter  $a^*$  were independent of frying time. Besides, Chroma parameter and color differences ( $\Delta E$ ) increased and Hue decreased significantly ( $P < 0.05$ ) as a function of frying time for  $t < 720$  s (Fig. 1b). During overcooking of the samples, all parameters remained constant.

### 3.4. Microstructure analysis of the coated and uncoated fried products

Scanning electron microscopy techniques (SEM and ESEM) were used to observe the structure of the fried dough. Fig. 2 shows that MC coating was getting dehydrated during the frying process and remained attached to the surface of the product, explaining the lower lipid content of the coated product. The thickness of the coating was measured on the micrographs obtaining values ranging between 9 and 24  $\mu\text{m}$  after 12 min of frying.

Fig. 2 also shows the integrity of the MC layer and the good adhesion of this coating to the fried product. The

addition of plasticizer (sorbitol) to MC coatings was necessary to achieve coating integrity (García et al., 2002). Formation of an uniform coating on the surface of the sample is essential to limit mass transfer during frying (Huse et al., 1998). The coating did not prevent the formation of a dehydrated zone on the surface of the dough.

The release of water vapor led to the formation of blisters at the outer surface of the crust being smaller in size and number in the coated doughs than in uncoated ones.

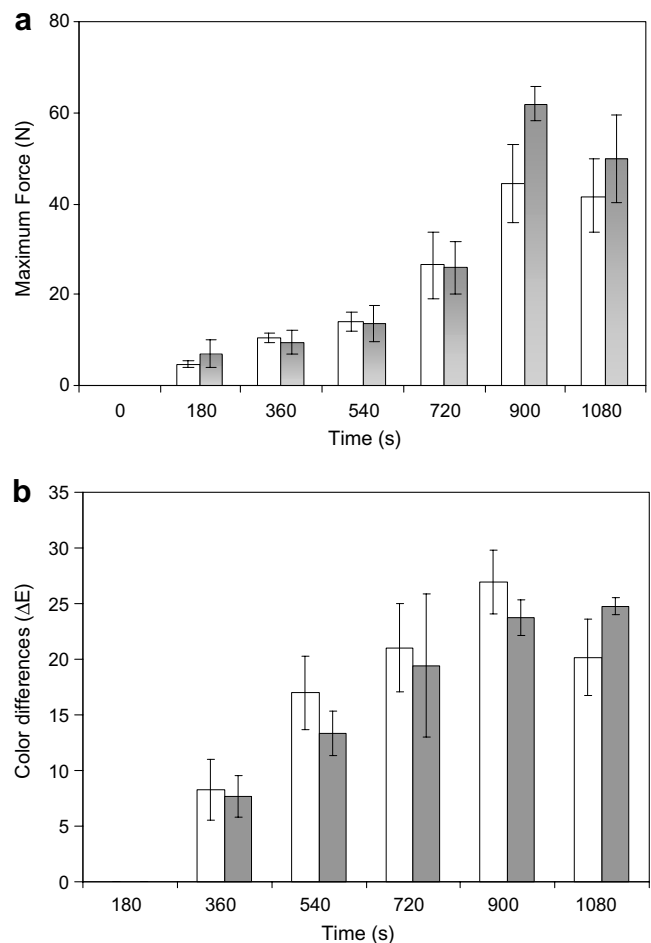


Fig. 1. Quality attributes of coated (filled bars) and uncoated (empty bars) fried dough discs as a function of frying time: (a) firmness; (b) color differences.

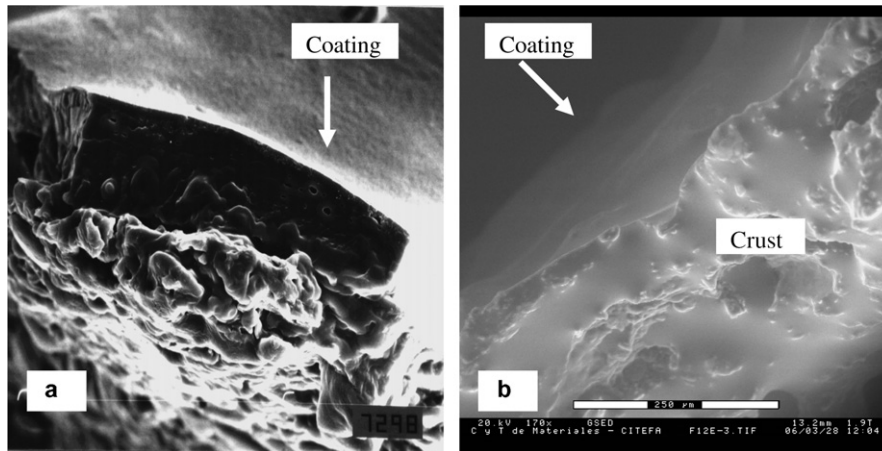


Fig. 2. Cross section micrographs of a fried dough disc coated with 1% methylcellulose plasticized with 0.75% sorbitol: (a) SEM, magnification 100 µm between marks; (b) ESEM.

In the first period of frying, the MC coating loses its water, the dough keeps its humidity and the starch gelatinizes with a higher water content than in uncoated doughs, leading to a more compact network (Fig. 3a and b).

During frying in both coated and uncoated systems, the core was progressively dehydrated and starch gelatinization was completed at 12 min.

### 3.5. Comparison of the experimental and simulated thermal histories and moisture content during frying

Experimental thermal histories of coated and uncoated samples did not differ significantly ( $P > 0.05$ ). Fig. 4a shows the measured values (symbols) at the center and in an inner point (1.6 mm from the center). After 120 s a constant temperature of 100.5 °C was reached at the core. The core temperature remained constant due to water evaporation while the temperature in the dehydrated zone increased up to the end of the process (15 min).

A cooling period could be observed when the product was removed from the fryer.

In the same Fig. 4a, simulated temperatures using the proposed model are shown (lines), and a good agreement with the experimental thermal histories is observed.

The presence of the coating did not change the moisture content of the samples during frying. Simulated and experimental water concentrations vs. frying times are shown in Fig. 4b and a good agreement was achieved. The assumption of a negligible barrier effect of the hydrophilic MC coating on water transfer was verified.

It must be emphasized that the mathematical model was only fed with properties obtained from the literature and that the tortuosity factor was the unique adjusted parameter in the water vapor effective diffusion coefficient. The value used in this work was 8.8, similar to that reported for several food and non-food products (Harper, 1962; Moldrup, Olesen, Komatsu, Schjonning, & Rolston, 2001).

The mathematical model was also validated on dough discs of 60 mm diameter and 5 mm thickness using the tortuosity value previously obtained; temperature profiles and water contents were satisfactorily predicted.

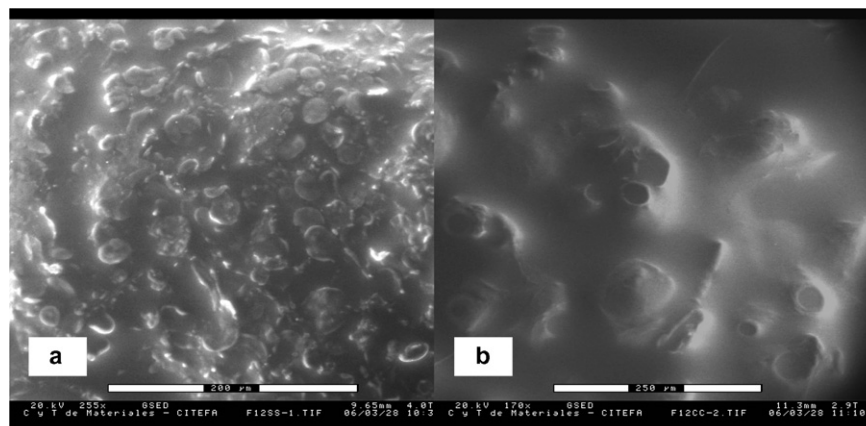


Fig. 3. ESEM micrographs of a dough disc fried during 12 min: (a) surface of the uncoated sample; (b) surface of the coated sample.

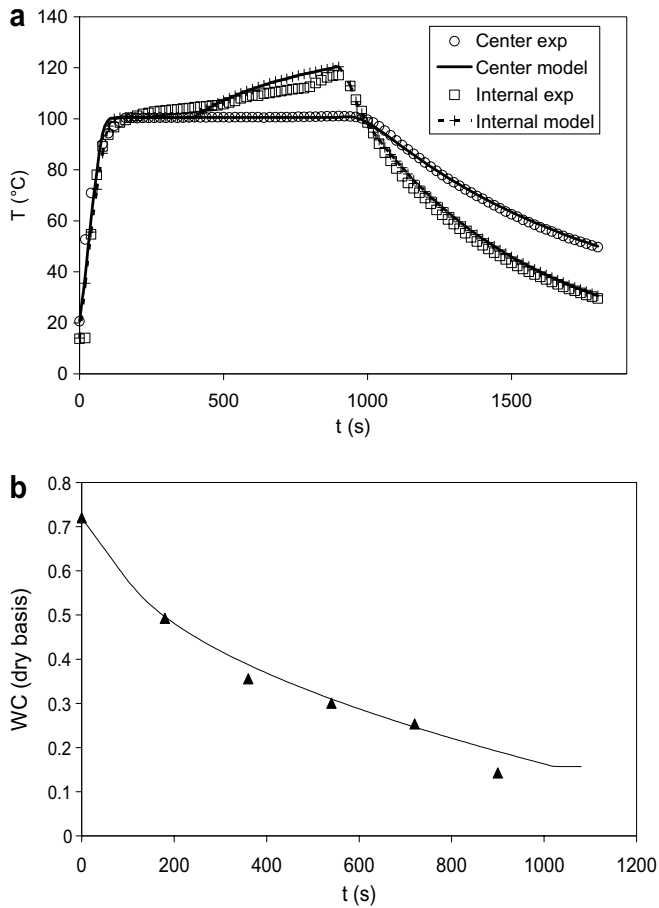


Fig. 4. (a) Thermal histories of the fried product at the center and in an internal point (1.6 mm from the border) along the different frying stages; (b) water content along the different frying stages. Experimental data (symbols) and numerical simulations (lines),  $L = 0.00357$  m,  $T_{ini} = 20$  °C.

Fig. 5a shows the predicted position of the vaporization front as a function of frying time. At 15 min of process, the vaporization front reached the center of the product, and the humidity corresponded to the bound water.

Water content correlated linearly with the position of the vaporization front  $x_1$  that corresponds to the thickness of the humidity core (Fig. 5b). Considering  $d_T = L - x_1$  the following equations were obtained :

$$WC = 0.14893 + 158.4 (L - d_T) \quad r^2 = 0.9966 \quad (24)$$

3.6. Relationship between oil uptake and microstructure changes during the frying process

Oil uptake depends on structural changes during the process; differences in the starting food microstructure can be expected to be important determinants in the evolution of the characteristics of the end product. Microstructural changes were produced during frying time; the dehydrated zone increased allowing the oil retained by the surface to penetrate into the pores formed by water evaporation.

The mathematical model solved in the present work allowed to estimate the thickness of the dehydrated zone ( $d_T = L - x_1$ ).

Fig. 6 shows oil uptake vs. frying time for both coated and uncoated samples. Simultaneously, the same figure shows the growth of the dehydrated zone with frying time. The predicted  $d_T$  (dehydrated zone thickness) curve as a function of time is the same for coated and uncoated samples, according to the proposed mathematical model.

A simple equation was proposed to interpret experimental results:

$$OU = a(1 - e^{-bt}) \quad (25)$$

where  $a$  is the oil concentration at long times and  $b$  is the coefficient that takes into account the structural changes as a function of frying time. The parameters of Eq. (25) were estimated by a non-linear regression showing high values of the correlation coefficient  $r^2$  (Table 3).

Results of Fig. 6 allowed to correlate oil uptake (OU) with the thickness of the dehydrated zone ( $d_T$ ) as shown in Fig. 7a. A linear behavior of oil uptake versus dehydrated zone thickness was maintained up to OU values of 0.031 (g/g dry solid) and 0.071 (g/g dry solid) for both coated and uncoated samples respectively.

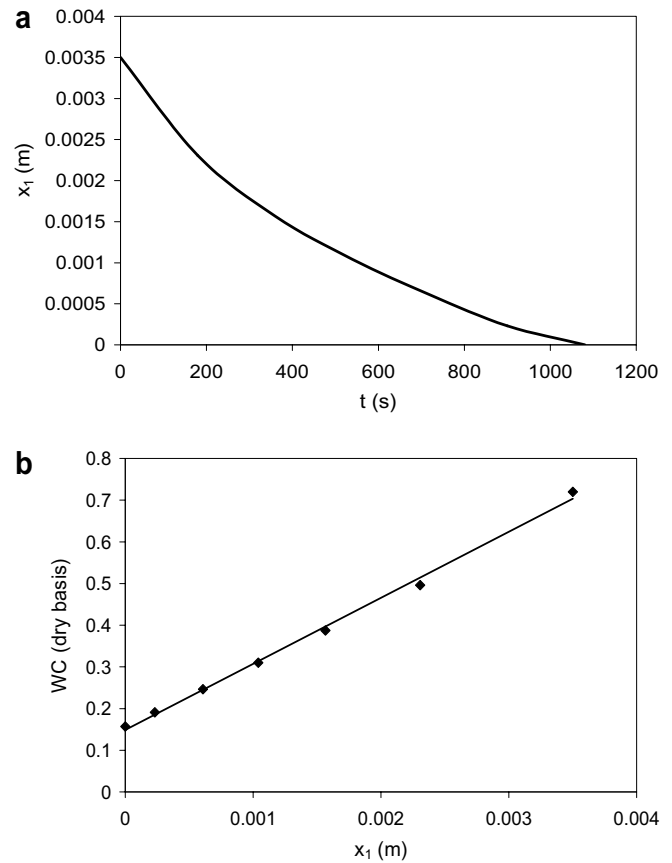


Fig. 5. (a) Position of the vaporization front as a function of frying time; (b) water content of fried dough discs as a function of the position of the vaporization front.



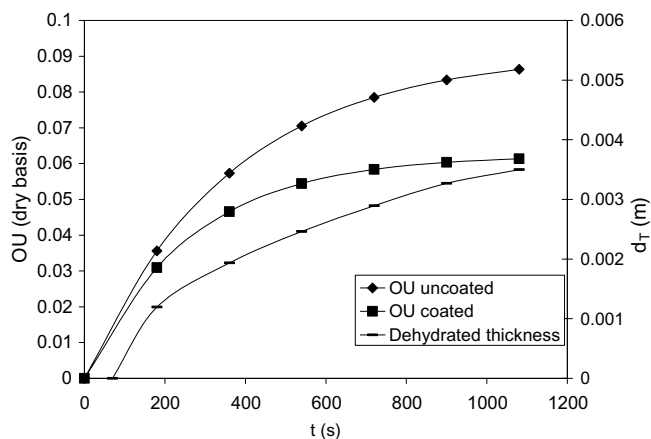


Fig. 6. Oil uptake of uncoated and coated samples and dehydrated zone thickness ( $d_T$ ) as a function of frying time.  $d_T$  curve is the same for coated and uncoated samples.

Table 3  
Oil absorption parameters of Eq. (25)

| Sample   | Parameter $a$ (g oil/g dry solid) | Parameter $b$ (1/s)   | Correlation coefficient ( $r^2$ ) |
|----------|-----------------------------------|-----------------------|-----------------------------------|
| Uncoated | 0.091                             | $2.76 \times 10^{-3}$ | 0.999                             |
| Coated   | 0.062                             | $3.81 \times 10^{-3}$ | 0.997                             |

The following linear regressions were obtained:

For uncoated samples:

$$OU = 28.86 d_T + 5 \times 10^{-4} \quad r^2 = 0.9991 \quad (26)$$

For coated samples:

$$OU = 25.931 d_T \quad r^2 = 1 \quad (27)$$

Values of OU for coated samples were lower than those of uncoated ones. For low  $d_T$  values, the oil retained by the surface could be incorporated into the dehydrated zone, when the sample was removed from the frying medium (linear relationship Fig. 7a). When the dehydrated zone was large, a deviation from the linear behavior was observed. This could be attributed to the fact that the amount of oil retained at the sample surface is limited, being the oil surface wetting the property related to the interfacial tension, governing these phenomena (Kassama, 2003).

The presence of MC coating with thermal gelation properties, modified the surface wetting and also became a mechanical barrier leading to a decrease in the OU of the coated samples.

The oil uptake was also correlated with the water content showing a negative slope. At high water contents (WC) the results were linearly correlated, however deviations were observed at WC = 0.3 and 0.35 (dry basis) for uncoated and coated samples, respectively.

Besides considering that WC is a function of  $d_T$  (Eq. (24)) the following linear equations were obtained for the initial frying periods (Fig. 7b):

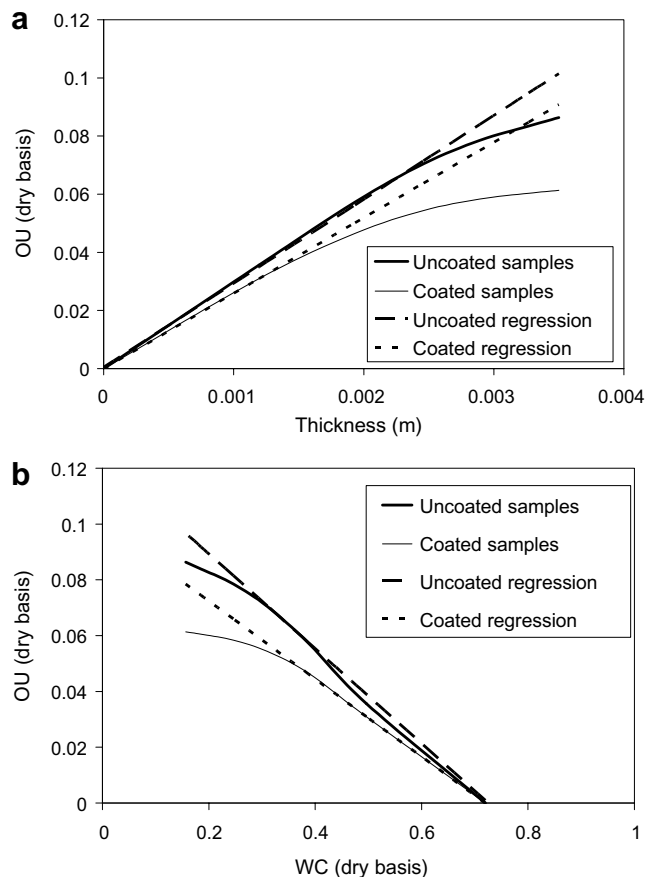


Fig. 7. Oil uptake of uncoated and coated samples as a function of: (a) dehydrated zone thickness; (b) water content of fried samples.

For the uncoated sample:

$$OU = -0.17WC + 0.1234 = 0.0981 + 26.928(d_T - L) \quad r^2 = 0.9978 \quad (28)$$

For the coated sample:

$$OU = -0.14WC + 0.1005 = 0.07965 + 22.176(d_T - L) \quad r^2 = 0.9999 \quad (29)$$

Following the oil uptake criterium introduced by Pinthus and Saguy (1994), the ratio between OU and WC was obtained from the aforementioned relationships, being 0.17 for uncoated and 0.14 for coated samples; these results agree with findings reported by Moyano and Pedreschi (2006).

#### 4. Conclusions

The application of MC coatings reduced significantly ( $P < 0.05$ ) the oil content of dough discs with regard to control ones, reaching a decrease of 30%. However, the coating did not modify the water content of the samples; this was attributed to the hydrophilic characteristics of the films that led to poor water vapor barrier properties, besides thermal histories of coated and uncoated samples were similar.

During the cooking process (for  $t < 720$  s) differences between the color parameters and firmness values of coated and uncoated samples were not significant; besides the panelists could not distinguish between control and coated samples.

Scanning electron microscopy techniques showed the integrity of the MC coating and good adhesion to the food product after its dehydration during deep oil frying process.

A mathematical model of the frying process for uncoated and coated samples based on the numerical solution of the heat and mass transfer differential equations under unsteady state conditions was proposed and solved using measured physical and thermal properties. It allowed to simulate satisfactorily the experimental data of temperature and water content during the different frying stages.

The model allowed to predict the position of the vaporization front and the thickness of the dehydrated zone as a function of frying time. Water content correlated linearly with the vaporization front position corresponding to the thickness of the humidity core.

Oil uptake (OU) that occurs when the sample is removed from the frying medium was correlated with the thickness of the dehydrated zone; a linear behavior was held for the initial frying period. However, deviations were observed when the thickness of the dehydrated zone increased. This could be attributed to the fact that the amount of oil retained at the sample surface is determined by the surface tension property. OU was also linearly correlated with water loss at the initial frying stage. A simple equation for oil uptake as a function of frying times was proposed, considering the microstructural changes developed during the frying process in which the dehydrated zone increases allowing the oil retained by the surface to penetrate into the pores left by water evaporation.

The presence of MC coating reduced the oil uptake due to the thermal gelation behavior, modifying the wetting properties and also becoming a mechanical barrier to the oil.

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