Research Article

Received: 20 January 2011 Revised: 15 July 2011 Accepted: 12 September 2011 Published online in Wiley Online Library:

(wileyonlinelibrary.com) DOI 10.1002/jsfa.4704

Plasticized methylcellulose coating for reducing oil uptake in potato chips

María José Tavera-Quiroz,[a] Marina Urriza,[a] Adriana Pinotti[a,b] and Nora Bertola[a]*

Abstract

BACKGROUND: As a result of consumers' health concerns and the trend towards healthier and low-fat food products, research has been undertaken to reduce the amount of fat absorbed in fried foods. This work focused on studying the efficacy of sorbitol and glycerol as plasticizers of methylcellulose coatings used to reduce oil uptake during the frying process of potato chips

RESULTS: Changes in color, mechanical properties, water activity and lipid oxidation during storage were monitored. Also, an explanation regarding the different performances between both methylcellulose coatings with and without plasticizer was attained and techniques from the field of packaging films such as dynamic mechanical analyzer (DMA) and Fourier transform infrared spectroscopy were applied to analyze the behavior of coatings submitted to the frying operation. The application of a methylcellulose coating was an adequate choice to reduce oil absorption in fried potato chips. The most effective formulation was 10 g L^{-1} methylcellulose with the addition of 7.5 g L^{-1} sorbitol. With the incorporation of this formulation, oil absorption was reduced by 30%. Neither the sorbitol concentration nor the presence of the MC coating affected the puncture maximum force and color parameters *L* and *a. The results of the sensory analysis indicated that the panelists could not distinguish between the coated and uncoated potato chips.**

CONCLUSION: Methylcellulose-based coating plasticized with sorbitol could be an alternative for obtaining healthier potato chips.

Keywords: potato chips; deep-fat frying; methylcellulose; coating; sorbitol; glycerol

INTRODUCTION

Potato chips are thin potato slices dehydrated by deep-fat frying up to a moisture content of 0.02 kg kg^{-1} or less. Deep-fat frying is widely used in industrial as well as institutional preparation of foods because consumers prefer the taste, appearance and texture of fried food products.[1–3] During the frying process mass transfer as well as heat transfer, takes place.[2] This is characterized by the movement of oil into the product as well as movement of water from the product into the oil.[4]

The use of edible films and coatings was proposed originally to extend the shelf life of meat and later to improve the quality of other fresh, frozen and manufactured food items. Polysaccharides, proteins, lipids and combinations of these have been used to produce edible coatings. When frying coated food pieces, the film hinders the absorption of the oil, improving its nutritional qualities and reducing the fat content and calories of the final product.[4–7] This application has become increasingly important in recent years, as oil uptake in fried products has turned into a health concern, related to obesity and coronary disease.[8–10] The properties of the food surface are very important for oil uptake, thus the application of a biopolymer coating is a promising route.[4,6,8] Among the hydrocolloids used for coating foods for frying, methylcellulose, a water-soluble polymer derived from cellulose by methylation, is also used as a pharmaceutical, cosmetic and food additive. Water-soluble methylcellulose solutions can form thermoreversible hydrogels in water on heating due mainly to a reduction of the hydrophobic association between methylcellulose chains and water at temperatures between 50 and 70 °C.[11,12] In the production of biopolymer-based films and coatings, plasticizers are essential additives since they can improve the flexibility and handling of films, maintain integrity and avoid pores and cracks in the polymeric matrix.[13] Plasticizers, which are low molecular weight components, increase the free volume between the polymer chains by reducing the number of active centers available for rigid polymer–polymer contacts or macromolecular mobility of the polymer. Compatibility between plasticizer and polymer is of major significance for effective plasticization and various parameters can indicate this feature, including polarity, hydrogen bonding, dielectric constant and solubility parameters.[14] Coating integrity is a critical factor related to adhesion and flexibility, decreasing possible discontinuities and brittle zones.[15–17] These characteristics are incompatible with the irregular shapes of most vegetables. In the case of frying applications, the integrity of the

* *Correspondence to: Nora Bertola, Centro de Investigación y Desarrollo en Criotecnología de Alimentos (CIDCA), CONICET La Plata, Facultad de Ciencias Exactas, 47 y 116, La Plata (1900), Argentina. E-mail: nbertola@cidca.org.ar*

a *Centro de Investigación y Desarrollo en Criotecnología de Alimentos (CIDCA), CONICET La Plata, Facultad de Ciencias Exactas, La Plata (1900), Argentina*

b *Facultad de Ingeniería, UNLP, La Plata (1900), Argentina*

coating also depends on the substrate; many foods increase their volume and change their shape after the frying. The addition of sorbitol as a plasticizer might facilitate the formation of a more effective coating at high temperatures, which allows decreasing the oil uptake.[15]

García *et al.*[18] noted that an edible methylcellulose coating plasticized with sorbitol on potato strips and dough discs caused an oil reduction of 40.6 and 35.2%, respectively. Likewise, a soy protein coating with the addition of glycerin was evaluated to reduce fat transfer during frying by Rayner *et al.*[19] Albert and Mittal[20] reported that a methylcellulose coating had some holes after frying and the addition of a plasticizer was necessary.

Abundant research has been carried out on the use of cellulose derivatives included in batters.[6,21–26] However, in the literature few studies have been reported on coatings of cellulose derivatives with and without the addition of plasticizers to reduce oil uptake to different food matrices,[10,20,27–30] and even fewer on fried potatoes.[4,18,31] These last studies were carried out on French fries.

As a consequence, this work had several aims. First, to study the efficacy of sorbitol and glycerol as plasticizers of methylcellulose coatings used to reduce oil uptake during the frying process of potato chips. Second, to monitor changes in color, texture, water activity and lipid oxidation through thiobarbituric acid reactive substances (TBARS) during the storage of these fried potatoes. Third, to attempt to find an explanation for the different performance between methylcellulose coatings with and without plasticizer, if it exists; and finally, to apply techniques from the field of packaging film such as dynamic mechanical analyzer (DMA) and Fourier transform infrared (FTIR) to analyze the behavior of coatings submitted to the frying operation.

MATERIALS AND METHODS

Potatoes (Spunta variety) and sunflower oil (AGD, General Deheza, Córdoba, Argentina) were used as materials. Potatoes were stored at 10 °C; after stabilization at room temperature for at least 24 h prior to use, the potatoes were peeled, washed and cut into slices of 0.18 ± 0.02 cm thickness using a Whirpool processor (Buenos Aires, Argentina). The center of each slice was removed with a 3.5 cm diameter punch. The samples were taken from the central medulla region of the potato, due to the existence of a more uniform cell structure. All of them were subjected to a blanching treatment at 80 °C for 5 min prior to the coating application, which was based on the work by Mai Tran[32] and on previous time and temperature screening.

Hydrocolloid solutions

Methylcellulose (A4M, Methocel) was provided by COLORCON S.A. (Buenos Aires, Argentina). To prepare the hydrocolloid solution, 1 g methylcellulose (MC) was slowly dispersed in 30 mL of hot distilled water (80 °C) under constant stirring for 1 h. Once a homogeneous system was obtained, total volume was completed to 100 mL with cold distilled water and the solution was kept under stirring until it reached room temperature.

Sorbitol (Merck, Gibbstown, NJ, USA) and glycerol (J.T. Baker, Xalostoc, Mexico) were added as plasticizers after MC had completely dissolved at different concentrations (2.5, 5.0, 7.5 and 10 g L^{-1} for sorbitol and glycerol) to select appropriate formulations for coating applications. Each experiment was carried out in triplicate. Solutions were prepared the day before frying and kept at room temperature. MC concentration was selected in previous work.[18]

Rheological characterization of hydrocolloid solutions

The rheological characterization of methylcellulose solutions 10 g L^{-1} (w/v) with and without plasticizer was performed in a Haake RheoWin 3.0 (Haake, Karlsruhe, Germany) rotational viscometer, at a controlled constant temperature of 25 °C. An MV I P type sensor system of roughened coaxial cylinders was used. Rheological curves were obtained after a stabilization time of 3 min at 25 °C. Shear stress (σ) was determined as a function of shear rate ($\dot{\gamma}$) between 0 and 500 s^{-1}, with the following program: 3 min to reach the maximum shear rate, then maintained for 1 min, and finally allowed to reach 0 shear rate in 3 min. Apparent viscosity was calculated at 500 s^{-1}.

Coating applications

Coatings of MC with and without plasticizer were applied by immersion of potato slices (approximately 8 g) in the prepared filmogenic solutions for 30 s. Then, samples were drained on a mesh to allow removal of the excess surface solution for 2 min and weighed to determine the amount of adhered coating. Each concentration of plasticizer was tested in triplicate.

Coating nomenclature used was for methylcellulose coating MC, methylcellulose coatings containing different plasticizer concentrations MC+2.5 P, MC+5 P, MC+7.5 P and MC+10 P, where P is sorbitol or glycerol.

Frying procedure

Coated and uncoated (control) potato slices were fried in a controlled temperature deep-fat fryer (Moulinex, Buenos Aires, Argentina) filled with 2 L of commercial sunflower oil. In each batch, between eight and ten potato slices were positioned on a metal mesh basket, immersed in the hot oil and fried. Standard frying conditions were 180 ± 2 °C for 2 min followed by blotting in tissue paper for 5 min. Used oil for frying was replaced by fresh oil after six frying batches. This fried time of 2 min was predetermined based on color and moisture considerations.

Moisture content and water activity

Moisture contents of coated and uncoated potato chips were determined by measuring their weight loss upon drying in an oven at 105 °C until reaching constant weight (dry sample weight). Results were expressed in grams of water per kilogram of dry potatoes.

Water activity of potato chips was evaluated by using an AquaLab Water Activity Meter (Decagon Devices, Inc., Pullman, Washington, USA) equipment. All the measurements were performed in triplicate.

Fat content

Around 2 g of potato chips with and without coating dried in an oven at 105 °C were placed in a thimble and the total crude fat was quantified with the Soxhlet method. Fat contents were determined in triplicate, with a mixture of petroleum ether–diethyl ether (1 : 1, v/v). The results were expressed in grams of fat per kilogram of dry potatoes.

Since the objective was to formulate a coating that reduces fat absorption, only the coating formulation that led to minimum oil content was selected for further studies.

Energy content

The caloric content of the potato chips was calculated using the Atwater values corresponding to lipids (9 kcal g^{-1}) considering the values of fat content. Results were expressed in kilocalories per 100 grams of potato chips.

Color measurements

The color of fried potatoes was determined with a Minolta colorimeter CR 400 Series (Osaka, Japan) calibrated with a standard ($Y = 93.2$, $x = 0.3133$, $y = 0.3192$). The CIELab scale was used; lightness (L) and chromaticity parameters a^* (red–green) and b^* (yellow–blue) were measured. Four potato chips of each batch were analyzed, recording three measurements for each potato chip.

Mechanical properties

Penetrometry tests were performed by using a 2 mm diameter cylindrical probe (SMSP/3) with a flat base at a constant rate of 5 mm s^{-1}. The potato chips were place on a platform with a 5 mm diameter hole. Tests were carried out in a texturometer TA.XT2i (Stable Micro Systems, Guildford, UK) with a 25 kg cell at room temperature. Each informed value corresponded at least to seven determinations. The curves of force (in newtons) as a function of the deformation (in millimeters) were recorded by Texture Expert Exceed software (Stable Micro Systems, Guildford, UK), and the maximum force was determined from these curves.

Water vapor resistance

The coated and uncoated fried potato slices were transferred to a desiccator placed in controlled room at 20 °C; previously the water activity of the potato chips was measured. To maintain a gradient across the system (potato matrix with and without coating), sodium chloride saturated solution (75% relative humidity) was used in the desiccator. Water vapor resistance was determined from the weight gain by the system. Changes in the weight of the system were recorded to the nearest 0.0001 g and plotted as a function of time. The diameter of the potato slices at initial and final control time was used to calculate the average surface area. The water vapor resistance (WVR) was estimated as follows:[33]

$$\mathrm{WVR} = \left\{ \frac{[(\%\mathrm{RH}/100) - a_w]}{RT} p_{wv} \right\} \frac{A}{J} \quad (1)$$

where % RH is the relative humidity of the desiccator, a_w the water activity of potato chips, p_{wv} is the water vapor pressure at 20 °C (mmHg), R is the universal gas constant (3464.629 mmHg cm^3 g^{-1} K^{-1}), T is the incubator temperature (K), A is the average surface area of potato slice (cm^2), and J is the slope of the weight gain curve (g s^{-1}).

Lipid oxidation

Lipid oxidation was measured by the TBARS test. The TBARS values were determined in duplicate on 2 g extract of fried potato sample according to Andrés *et al.*[34] to evaluate the extent of oxidative rancidity development. Two independent extracts were obtained for each sample and subjected to the thiobarbituric acid reaction for 30 min at 70 °C, followed by a cooling for 30 min at 0 °C. Determinations were performed in duplicate, by using a spectrophotometer (Spectrophotometer DR/2000 HACH, Loveland, USA) to determine the absorbance of the sample at 532 nm. Results were expressed as mg malonaldehyde per kg.

Sensory analysis

Fried potato samples with and without coating were evaluated by a non-trained sensory panel of 22 members about 10 min after their preparation. A triangle test was performed to determine if consumers could distinguish between coated (using the formulation that led to minimum oil content) and uncoated samples. Each group of samples of the triangle test, consisting of three chips, was randomly numbered and presented to the panel member with the instructions to taste the samples from left to right and to pick out the two equal samples from the three. The trial was replicated twice.

Storage accelerated test

An accelerated testing to monitor the quality of fried potato chips uncoated and coated was carried out; hence batches from 10 to 12 potato slices approximately (with and without coating) were fried and then packaged under air atmosphere in containers independently formed from the biaxial oriented polypropylene film. Samples were maintained in a controlled room at 20 °C and 60% RH. Each sample was extracted twice and each extraction analyzed by duplicated for different times (15, 30, 60 and 90 days). During the storage, color, texture, water activity and TBARS measurements were carried out according to the procedures aforementioned.

Film preparation

Because the properties of a coating on the surface of a fried biological matrix cannot be characterized, isolated films have been reported as an alternative for predicting coating properties.[35] Thus, MC films were prepared as model systems.

About 20 g of solutions with 7.5 g L^{-1} sorbitol and without plasticizer were cast onto Petri dishes (9 cm diameter) and dried at 37 °C in an oven until constant weight. The films obtained were removed from the dish and then were stored at 20 °C and 60% RH in a controlled room prior to tensile and water vapor permeability tests.

On the other hand, MC solutions were also deposited on an inert ceramic material that was submitted to the frying operation to obtain a partially detachable coating, which, once detached, was used as a sample to obtain the corresponding spectra by FTIR.

Film thickness was determined using a coating thickness gauge Check Line DCN-900 (Cedarhurst, New York, USA) for non-conductive materials on non-ferrous substrates. The informed values correspond to the average of at least 15 measurements at different positions for each specimen.

Film stress–strain behavior

Probes of 6 mm × 30 mm were used to analyze the tensile stress–strain behavior of the MC films with and without plasticizer through a dynamic mechanical analyzer DMA TA Instruments-Q 800 (New Castle, DE, USA), equipped with tension clamps. One end of the strand was attached to a superior mobile clamp and the other end attached to a lower fixed clamp. For quasi-static test in uniaxial condition, a preload force of 0.2 N and a constant force ramp rate 0.2 N min^{-1} were applied to record the stress–strain curves until reaching 18 N or the film rupture. Tests were carried out at 100 and 180 °C; these temperatures were selected considering the vapor transfer temperature during the frying operation and the maximum temperature reached by the system. At least five replicates of each condition were obtained. According to Mancini

et al.[36] and Del Nobile *et al.*[37] a mathematical model capable of describing the entire stress–strain curve is:

$$\sigma_T = E_C \varepsilon_T \exp(\varepsilon_T K) \quad (2)$$

where ε_T and σ_T (MPa) are the true strain and the true stress, respectively, E_C (MPa) is the elastic modulus (the tangent to the stress–strain curve at the origin), and K is a constant considered as a fitting parameter.

Film water vapor permeability

Water vapor permeability (WVP) tests of the films were conducted using the ASTM method E96 with several modifications as described in previous work.[17] Each film sample was sealed over a circular opening of 0.00181 m^2 in a permeation cell that was stored at 20 °C in a dessicator. To maintain the driving force corresponding to a 75% RH gradient across the film, anhydrous calcium chloride (0% RH) was placed inside the permeability cell and a sodium chloride saturated solution (75% RH) was used in the desiccator. Eight weight measurements were made for about 10 h. Slopes were calculated by linear regression and correlation coefficients (r^2) for reported data were 0.99 or greater.

Fourier transform infrared spectroscopy of films and coatings

The FTIR spectra of the films and fried coatings were recorded in an IR spectrometer (model Nicolet iS10; Thermo Scientific, Madison, WI, USA) in the wavenumber range 4000 to 400 cm^{-1} by accumulation of 64 scans at 4 cm^{-1} resolution.

Statistical analysis

Analysis of variance (ANOVA) using the general linear model was carried out to evaluate the levels of significance of the analyzed factors, using SYSTAT software version 10.0 (SYSTAT Inc., Evanston, IL, USA). The least square difference test (LSD) was used for mean comparisons with $P < 0.05$.

RESULTS AND DISCUSSION

Hydrocolloid solutions

The rheological study of the filmogenic solutions is important because if the viscosity is excessively high, the application of a coating might be difficult or even impossible. Conversely, if the viscosity is too low the coating will not adhere to the potato tissue, and will have a tendency to fall off. Consequently, the solution viscosities are related to the spreading and the covering capacities of the coatings. However other properties influence on the adherence of the coating. Varela and Fiszman[7] reported that MC and HPMC thermal gelling ability encourages cohesion and adherence to the surface of the substrate.

The apparent viscosity value of MC solution (0.102 Pa s at 500 s^{-1}, about 100 times water viscosity) turned out to be similar to those obtained by García *et al.*[18] at 691 s^{-1}. The addition of sorbitol did not modify the apparent viscosity of the MC solution without plasticizer regardless of the concentration used (Table 1). On the other hand, the apparent viscosity of solutions with increasing concentrations of glycerol did not show significant differences ($P > 0.05$) among them, but it did differ with respect to the MC solution.

The amount of adhered solution is shown in Table 1. Sorbitol exhibited higher adhesion than glycerol; this result would allow a prediction of the formation of a more effective coating during the frying process. According to Viña *et al.*[38] the addition of a plasticizer decreased surface tension, facilitating coating adhesion to foodstuffs. Itoh *et al.*[39] have suggested that sorbitol may be acting as a dehydration agent in a similar manner to 'salting-out' of electrolytes such as NaCl, which enhance the thermal aggregation of methylcellulose.

Table 1. Apparent viscosities determined at 500 s^{-1} and weight of adhered solution of methylcellulose solutions, 10 g L^{-1} (w/v), with and without plasticizer (glycerol or sorbitol)

Concentration of plasticizer (g L^{-1} w/v)	Apparent viscosity at 500 s^{-1} (Pa s)		Weight of adhered solution (g $g^{-1}_{potatoes}$)	
	Glycerol	Sorbitol	Glycerol	Sorbitol
0.0	0.102[a]	0.102[a]	0.33[a]	0.33[a]
2.5	0.075[b]	0.114[a]	0.41[b]	0.72[c]
5.0	0.074[b]	0.115[a]	0.46[b]	0.79[c]
7.5	0.071[b]	0.098[a]	0.47[b]	0.74[c]
10.0	0.071[b]	0.115[a]	0.46[b]	0.68[c]

[a,b,c] Means within a row without a common superscript differ ($P < 0.05$).

The effect of coating and storage on the properties of potato chips

The application of MC coatings reduced oil absorption and increased water retention in the samples of potato chips (Fig. 1a and b). The increase of the glycerol concentration not only failed to improve the oil barrier properties of the coating, but it also led to a greater amount of oil uptake with the addition of 7.5 g L^{-1} (Fig. 1b). In contrast to this behavior, the addition of sorbitol enhanced the barrier properties of the coatings.

Sorbitol is a polyhydric alcohol with a high boiling point. It is water-soluble, polar and non-volatile.[40,41] Permanence is related to volatility and resistance to migration and extraction in water, solvents and oils. Therefore, the behavior of the sorbitol could be related to the low vapor pressure and the low rate of diffusion in the polymer.[16]

According to the results obtained in the analysis of variance, sorbitol 7.5 g L^{-1} was the most effective concentration, thus this one was selected for further studies. The application of this formulation reduced the oil uptake by 30% with respect to uncoated samples during the deep fat-frying. It could be attributed to the fact that the plasticizer reduced flaking and cracking by improving coating flexibility and toughness as it was aforementioned.

García *et al.*[18] reported that MC coating formulations were the most effective, reducing oil-uptake by 35–40% depending on the product. Besides barrier properties, flexibility of the coating through a plasticizer is a required coating characteristic because sample volume frequently changes during frying and coating integrity may be compromised.

Williams and Mittal[27] found that MC coatings (20 g L^{-1}) reduced the lipid content of potato spheres by 34.5%. Aminlari *et al.*,[42] using potato chips with a protein-based coating, reported a reduction of fat between 5 and 14%. On the other hand, Khalil,[31] working on French fried potatoes coated with pectin found a reduction in oil content compared to control from 1.2 to 16.0%, respectively. The difference can be attributed to the different coating formulation, frying medium and product used.

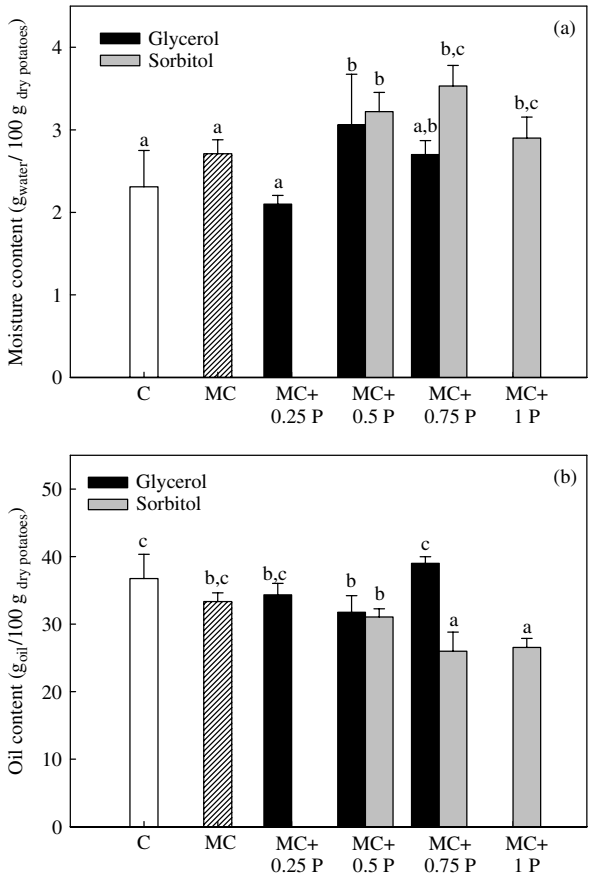

Figure 1. Moisture content (a) and oil content (b) of potato chips with and without coating. C, control (without coating); MC, 10 g L^{-1} methylcellulose; P, plasticizer (sorbitol or glycerol). [a,b]Different letters indicate significant differences ($P < 0.05$).

Table 2. Caloric content of potato chips control (C) and coated with 10 g L^{-1} methylcellulose without (MC) and with addition of sorbitol (MC+S)

Sample	Caloric content (kcal 100 $g^{-1}_{potato\ chips}$)
C	322.9^{c}
MC	$291.8^{b,c}$
MC + 5 S	$271.6^{a,b}$
MC + 7.5 S	226.0^{a}
MC + 10 S	231.8^{a}

[a,b,c] Means without a common superscript differ ($P < 0.05$).

Table 3. Color parameters, maximum force and water vapor resistance (WVR) obtained for potato chips control (C) and coated with 10 g L^{-1} methylcellulose without (MC) and with addition of 7.5 g L^{-1} sorbitol (MC+7.5 S)

Sample	L	a^*	b^*	Maximum force (N)	WVR (s cm^{-1})
C	66.26^{a}	1.09^{a}	34.26^{b}	1.26^{a}	59.28^{a}
MC	65.34^{a}	1.17^{a}	30.06^{a}	1.34^{a}	80.76^{b}
MC + 7.5 S	67.49^{a}	1.00^{a}	30.91^{a}	1.49^{a}	118.78^{c}

[a,b,c] Means within a row without a common superscript differ ($P < 0.05$).

Considering the results of water retention, the moisture of the control was 23 g $kg^{-1}_{dry\ potatoes}$ whereas the highest value corresponded to coated samples with MC+7.5 S with a value of 35 g $kg^{-1}_{dry\ potatoes}$. Aminlari *et al.*[42] attempted to explain the mechanism by which coatings work would be to reduce water loss during frying which, in turn, would decrease the absorption of oil. Bertolini Suárez *et al.*[30] found that 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 oil.

Methylcellulose film plasticized by glycerine provided a fat reduction of 58.2% and a water loss decrease of 26.4% according to the report of Albert and Mittal[20] working on dough.

The oil content obtained for potato chips with and without coating was used to calculate the caloric content of the samples taking the conversion factor from kcal-fat (9 kcal g^{-1}_{fat}) into account; the results are shown in Table 2. The caloric content of fat of the control samples (322.9 kcal 100 $g^{-1}_{potato\ chips}$) was in agreement with the highest oil content. In comparison, commercial potato chips add between 300 and 350 kcal 100 g^{-1} based on fat content. The caloric content of potato chips coated with MC+7.5 S was decreased by 30% with respect to the control and by 22.5% in relation to samples coated with MC, in accordance with the decrease in oil uptake.

Surface color measurements were conducted on control and coated fried samples with 10 g L^{-1} MC with and without the addition of 7.5 g L^{-1} sorbitol (Table 3). Non-significant differences ($P > 0.05$) were observed in the values of both luminosity (L) and chromaticity parameter a^* of the potato chips with and without coating regardless of the sorbitol presence. CIE b^* values of coated fried samples underwent a decrease with respect to those of uncoated potato chips. However, the addition of 7.5 g L^{-1} sorbitol to MC solutions did not alter the above-mentioned parameter values (Table 3).

Similar surface color parameters were informed by Romani*et al.*[9] working on potato chips at 180 °C and García *et al.*[18] on French fries at 180 °C.

Other properties were also studied on MC coating with and without the addition of 7.5 g L^{-1} sorbitol on fried potatoes formulation compared with the control. Texture is a fundamental property in potato chips and it is basically influenced by water content. In the just-fried potato chips, notwithstanding the differences in the moisture content of the samples with and without coating, the analysis of variance showed that neither the presence of the plasticizer nor its concentration in the coating affected the maximum force value significantly ($P > 0.05$), as can be seen in Table 3. This is a favorable result because it is desired that the coating does not have a significant impact on the texture of fried products. Similar force values were obtained by Pedreschi and Moyano[43] working on potato chips and by Rayner *et al.*[19] using soy protein-based films on disks of potato chips. Furthermore, Funami *et al.*,[44] working on doughnuts, found that MC addition had no effect on breaking stress of the samples.

All just-fried potatoes coated with and without sorbitol in the formulation, showed initial higher value of water activity (a_w) than the uncoated samples (control), as can be seen in Fig. 2a.

In order to match the results obtained with instrumental measurements a sensory analysis was performed on the just-fried samples of potato chips with and without coatings. The results of this analysis showed that 88% of the panelists found no differences between these samples.

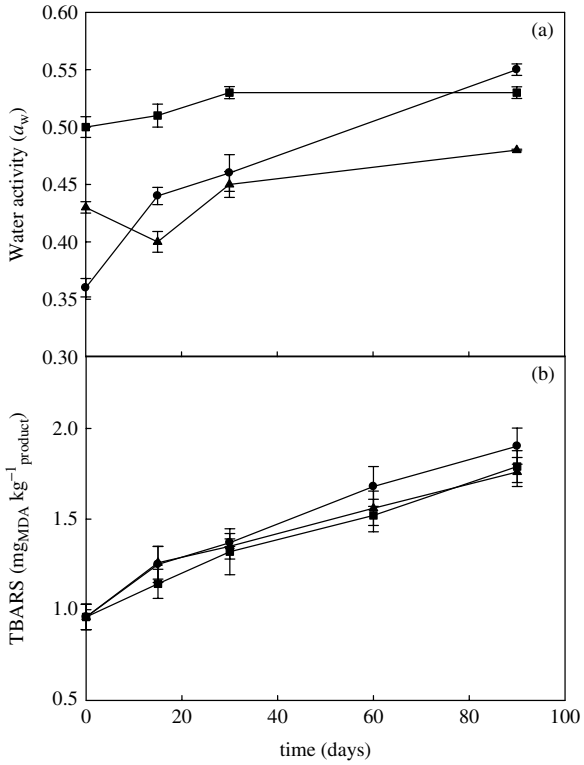

Figure 2. Changes in (a) water activity (a_w) and (b) thiobarbituric acid reactive substance (TBARS) values of potato chips (●) control, (▲) coated with 10 g L^{-1} MC and (■) coated with MC + 7.5 S, during storage time.

During the storage time color parameter b^*, water activity, texture and lipid oxidation were monitored. The values of the parameter b^* were maintained with significant differences ($P < 0.05$) among uncoated and coated potato chips. Nevertheless, there was no difference ($P > 0.05$) along the storage time for each kind of sample.

Water activity values of the coated samples remained constant. Nevertheless, a_w underwent an increase in the case of uncoated samples, reaching similar value to those found for samples coated with MC after 30 days of storage. After 90 days, it became higher than coated potato chips with and without plasticizer (Fig. 2a).

In spite of the water activity of uncoated samples matched with those values obtained for coated chips, there was no significant ($P > 0.05$) variation in the maximum force values. These results would be in accordance with those observed by Katz and Labuza,[45] who pointed out that crispy snacks have a critical water content. At low water activities the water content is insufficient to affect mechanical properties.

In the case of the TBARS test, the measured values as indicators of lipid oxidation increased during the storage and with it the smell and taste rancid, but the increase occurred in both types of samples, control and coated. Hence, no significant differences between samples ($P > 0.05$) were observed (Fig. 2 b). Air is known to be a pro-oxidant agent; consequently the results achieved with this atmosphere were expected.[46]

Barrier properties of model films in relation to fried potato chips

MC films with and without sorbitol were homogeneous, thin, and easy to handle in all concentrations assayed. The films were removed without difficult from the acrylic plates. They had thickness values between 20 and 25 μm. WVP results obtained for MC films with and without sorbitol did not differ significantly ($P > 0.05$), ranging between 6.7 and 5.7 10^{-11} g m^{-1} s^{-1} Pa^{-1} (Table 4). Similar values were found by Piermaría *et al.*[17] working on kefiran films plasticized with sorbitol. Instead, water vapor resistance (WVR, Equation 1), which was studied on coated and uncoated just-fried potatoes, showed significant differences ($P < 0.05$) between samples (Table 3); the addition of sorbitol led to higher WVR. The WVR value in the case of potatoes coated with MC+7.5 S was twice that obtained for uncoated samples and 36% higher than that found for potatoes coated with MC.

Table 4. Water vapor permeability and elastic modulus obtained for film with 10 g L^{-1} methylcellulose without (MC) and with addition of 7.5 g L^{-1} sorbitol (MC+7.5 S)

Sample	Water vapor permeability (g Pa^{-1} m^{-1} s^{-1})	Elastic modulus, E_c (MPa) 100 °C	180 °C
MC	5.74×10^{-11}[a]	311.90[a]	38.59[c]
MC+7.5 S	6.67×10^{-11}[a]	64.98[b]	5.10[d]

[a,b,c,d] Different letters indicate significant differences ($P < 0.05$).

The different behavior between films and coatings could be explained by considering that the coating without sorbitol turned out to be a rigid matrix, which did not allow the total area of the potato slices to be covered during the frying process. That is, the formation of pores or cracks permitted the transfer of a major amount of water vapor. This fact was not observed in model films prepared for determining WVP, which showed a uniform surface without pores or cracks, independently of the presence of the plasticizer.

Film stress–strain tests

The tensile stress–strain curves of the MC films with and without plasticizer are shown in Fig. 3, in which experimental data were fit with those predicted by Equation 2. Statistical analysis indicated that a good agreement was obtained for film of MC and MC containing 7.5 g L^{-1} sorbitol ($r^2 > 0.98$). The mechanical behavior of unplasticized films exhibited higher values of stress and lower values of strain than plasticized films at both temperatures 100 and 180 °C; the higher the temperature the higher the strain, for MC as well as MC+7.5 S. Similar behavior was observed by Gupta *et al.*[47] working on PET at different temperatures and León *et al.*[48] on a gellan matrix. Table 4 exhibits the elastic modulus obtained from the Equation 2 for all samples assayed; the elastic modulus (E_C) decreased with the increase of temperature and with the addition of the plasticizer. As suggested from the dynamic results obtained, the addition of sorbitol as plasticizer might facilitate macromolecular mobility, increasing the elongation and the formation of a more effective coating at high temperatures, which allowed decreasing the oil uptake.

Fourier transform infrared spectra of the coatings

Figure 4 shows the spectra of plasticized and unplasticized MC films and fried MC coatings. The unplasticized MC spectra of films exhibited absorption bands at 3600–3000 cm^{-1} with a minimum at 3470 cm^{-1} owing to the O–H stretching, 2920 cm^{-1} (C–H stretching), 1640 cm^{-1} (C–O) and 1066 cm^{-1} (C–O–C) (Fig. 4a). These results are in accordance with those observed by Zaccaron

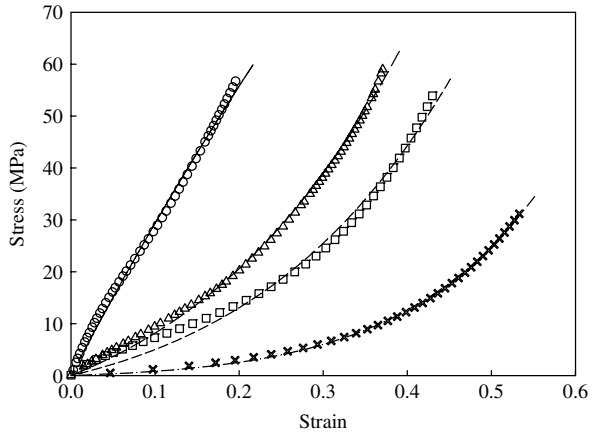

Figure 3. Stress–strain behavior of films formulated with (○) MC 100 °C; (———) model MC 100 °C; (△) MC + 7.5 S 100 °C; (– – –) model MC + 7.5 S 100 °C; (□) MC 180 °C; (- - - -) model MC 180 °C; (×) MC + 7.5 S 180 °C; (— · — · —) model MC + 7.5 S 180 °C.

et al.[49] In the spectra of plasticized MC films, the previously mentioned region, 3600–3000 cm^{-1}, became broader due to the water absorption from the material (Fig. 3b). The region from 1270 to 1500 cm^{-1} corresponds to vibration modes of methylcellulose groups, and the region from 1580 to 1700 cm^{-1} corresponds to the bending mode of water molecules contained in the film,[50] the latter being broader for plasticized MC films. According to Filho *et al*.[51] the distinctive absorption bands at 950 cm^{-1}, characteristic of the methylation of methylcellulose was evident in MC as well as in MC+7.5 S samples. Instead, the band located at wavenumber 900 cm^{-1} referred to the C–O–C of the pyranose ring was only observed in plasticized film spectra.[52]

In the case of fried coating spectra (Fig. 4a and b), the location of the bands was similar, but the peak intensities was lower than those of corresponding films, indicating a thickness much smaller. The principal feature was the appearance of a peak at 1740 cm^{-1}, which has been ascribed to the stretching vibration of the ester carbonyl functional group of the triglycerides in edible oils, and also in lipids in pork adipose tissue and farmed salmon.[53,54]

The results obtained were in agreement with the fact that the application of plasticized MC coatings on potato chips led to higher water content in relation to unplasticized MC coatings.

CONCLUSIONS

The application of a MC coating plasticized with sorbitol was an effective choice to reduce oil absorption in fried potato chips. From the results obtained by instrumental measurements and sensory panel, it was possible to optimize a MC formulation containing a plasticizer which decreased the incorporation of oil in potato chips during the frying operation. Consumer acceptance was similar to that obtained with the control product since they were not able to distinguish among them. Films used as models allowed the explaination that the presence of sorbitol improved the flexibility and mechanical properties of the films and their integrity, through DMA and FTIR. Due to the impact of snacks on people's food habits and the trend for consumers to re-evaluate the nutritional importance of food, methylcellulose-based coatings could be an alternative for obtaining healthier potato chips.

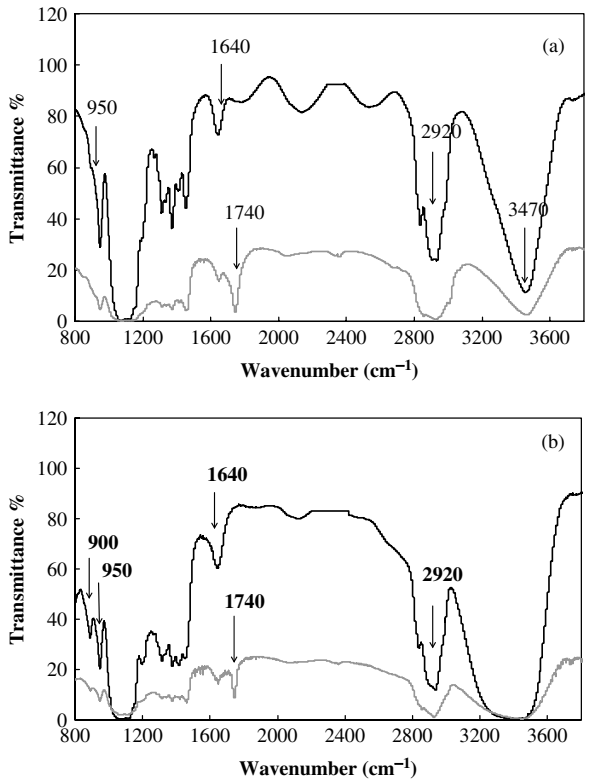

Figure 4. FTIR spectra of (a) MC film (upper spectrum) and fried coating (lower spectrum), and (b) MC+7.5 S film (uppper spectrum) and fried coating (lower spectrum).

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support provided by Agencia Nacional de Promoción Científica y Técnica (ANPCyT), Universidad Nacional de La Plata (UNLP) and Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET, La Plata) of Argentina.

REFERENCES

1 Garayo J and Moreira R, Vacuum frying of potato chips. *J Food Eng* **55**:181–191 (2002).
2 Mellema M, Mechanism and reduction of fat uptake in deep fat fried foods. *Trends Food Sci Technol* **14**:364–373 (2003).
3 Moyano PC and Pedreschi F, Kinetics of oil uptake during frying of potato slices: Effect of pre-treatments. *LWT–Food Sci Technol* **39**:285–291 (2006).
4 Rimac-Brnčić S, Lelas V, Rade D and Šimundić B, Decreasing of oil absorption in potato strips during deep fat frying. *J Food Eng* **64**:237–241 (2004).
5 Balasubramaniam VM, Chinnan MS, Mallikarjunan P and Phillips RD, The effect of edible film on oil uptake and moisture retention of a deep-fat fried poultry product. *J Food Process Eng* **20**:17–29 (1997).
6 Salvador A, Sanz T and Fiszman SM, Performance of methyl cellulose in coating batters for fried products. *Food Hydrocolloids* **22**:1062–1067 (2008).
7 Varela P and Fiszman SM, Hydrocolloids in fried foods. A review. *Food Hydrocolloids* **25**:1801–1812 (2011).
8 García M, Bifani V, Campos C, Martino MN, Sobral P, Flores S, *et al*., Edible coating as an oil barrier or active system, in *Food Engineering: Integrated Approaches*. *Food Engineering Series*. Springer, New York, pp. 225–241 (2008).
9 Romani S, Rocculi P, Mendoza F and Dalla Rosa M, Image characterization of potato chip appearance during frying. *J Food Eng* **93**:487–494 (2009).

10 Quasem JM, Mazahreh AS, Abu-Alruz K, Afaneh IA, Al-Muhtaseb AH and Magee TRA, Effect of methylcellulose coating and pre-treatment on oil uptake, moisture retention and physical properties of deep-fat fried starchy dough system. *Am J Agric Biol Sci* **4**:156–166 (2009).
11 Li L, Shan H, Yue CY, Lam YC, Tam KC and Hu X, Thermally induced association and dissociation of methylcellulose in aqueous solutions. *Langmuir* **18**:7291–7298 (2002).
12 Hatakeyama H, Onishi T, Endo T and Hatakeyama T, Gelation of chemically cross-linked methylcellulose studied by DSC and AFM. *Carbohydr Polym* **69**:792–798 (2007).
13 Donhowe IG and Fennema OR, The effects of plasticizers on crystallinity, permeability, and mechanical properties of methylcellulose films. *J Food Process Preserve* **17**:247–257 (1993).
14 Vieira MGA, da Silva MA, dos Santos LO and Beppu MM, Natural-based plasticizers and biopolymer films: A review. *Eur Polym J* **47**:254–263 (2011).
15 Krochta J, Baldwin EA and Nisperos-Carriedo M, *Edible Coating and Films to Improve Food Quality*. Technomic Publishing Company, Lancaster, USA (1994).
16 Mohsin M, Hossin A and Haik Y, Thermomechanical properties of poly(vinyl alcohol) plasticized with varying ratios of sorbitol. *Mater Sci Eng A* **528**:925–930 (2011).
17 Piermaria J, Bosch A, Pinotti A, Yantorno O, Garcia MA and Abraham A, Kefiran films plasticized with sugars and polyols: Water vapor barrier and mechanical properties in relation to their microstructure analyzed by ATR/FT-IR spectroscopy. *Food Hydrocolloids* **25**:1261–1269 (2011).
18 García MA, Ferrero C, Bertola N, Martino M and Zaritzky N, Edible coatings from cellulose to reduce oil uptake in fried products. *Innov Food Sci Emerg Technol* **3**:391–397 (2002).
19 Rayner M, Ciolfi V, Maves B, Stedman P and Mittal GS, Development and application of soy-protein films to reduce fat intake in deep-fried foods. *J Sci Food Agric* **80**:777–782 (2000).
20 Albert S and Mittal G, Comparative evaluation of edible coatings to reduce fat uptake in a deep-fried cereal product. *Food Res Int* **35**:445–458 (2002).
21 Fiszman SM and Salvador A, Recent developments in coatings batters. *Trends Food Sci Technol* **14**:399–407 (2003).
22 Sanz T, Salvador A and Fiszman SM, Effect of concentration and temperature on properties of methylcellulose-added batters. Application to battered, fried seafood. *Food Hydrocolloids* **18**:127–131 (2004).
23 Llorca E, Hernando I, Perez-Munuera I, Quiles A, Larrea V, Fiszman SM, *et al*., Microstructural study of frozen batter-coated squid rings prepared by an innovative process without a pre-frying step. *Food Hydrocolloids* **19**:297–302 (2005).
24 Sahin S, Sumnu SG and Altunakar B, Effects of batters containing differentgum types on the quality of deep-fat fried chicken nuggets. *J Sci Food Agric* **85**:2375–2379 (2005).
25 Akdeniz N, Sahin S and Sumnu G, Functionality of batters containing different gums for deep-fat frying of carrot slices. *J Food Eng* **75**:522–526 (2006).
26 Albert A, Perez-Munuera I, Quiles A, Salvador A, Fiszman SM and Hernando I, Adhesion in fried battered nuggets: Performance of different hydrocolloids as predusts using three cooking procedures. *Food Hydrocolloids* **23**:1443–1448 (2009).
27 Williams R and Mittal GS, Water and fat transfer properties of polysaccharide films on fried pastry mix. *LWT–Food Sci Technol* **32**:440–445 (1999).
28 Holownia KI, Chinnan MS, Erickson MC and Mallikarjunan P, Quality evaluation of edible film-coated chicken strips and frying oils. *J Food Sci* **65**:1087–1090 (2000).
29 Maskat MY, Yip HH and Mahali HM, The performance of a methyl cellulose-treated coating during the frying of a poultry product. *Int J Food Sci Technol* **40**:811–816 (2005).
30 Bertolini Suárez R, Campañone L, García MA and Zaritzky N, Comparison of the deep frying process in coated and uncoated dough systems. *J Food Eng* **84**:383–393 (2008).
31 Khalil AH, Quality of French fried potatoes as influenced by coating with hydrocolloids. *Food Chem* **66**:201–208 (1999).
32 Mai Tran TT, Chen XD and Southern C, Reducing oil content of fried potato crisps considerably using a 'sweet' pre-treatment technique. *J Food Eng* **80**:719–726 (2007).
33 Villalobos-Carvajal R, Hernández-Muñoz P, Albors A and Chiralt A, Barrier and optical properties of edible hydroxypropyl methylcellulose coatings containing surfactants applied to fresh cut carrot slices. *Food Hydrocolloids* **23**:526–535 (2009).
34 Andrés S, Zaritzky N and Califano A, Innovations in the development of healthier chicken sausages formulated with different lipid sources. *Poultry Sci* **88**:1755–1764 (2009).
35 Ye Z-w, Rombout P, Remon J R, Vervaet C and Van den Mooter G, Correlation between the permeability of metoprolol tartrate through plasticized isolated ethylcellulose/hydroxypropyl methylcellulose films and drug release from reservoir pellets. *Eur J Pharm Biopharm* **67**:485–490 (2007).
36 Mancini M, Moresi M and Rancini R, Mechanical properties of alginate gels: Empirical characterization. *J Food Eng* **39**:369–378 (1999).
37 Del Nobile MA, Chillo S, Mentana A and Baiano A, Use of the generalized Maxwell model for describing the stress relaxation behavior of solid-like foods. *J Food Eng* **78**:978–983 (2007).
38 Viña SZ, Mudridge A, García MA, Ferreyra RM, Martino MN, Chaves AR, *et al*., Effects of polyvinylchloride and edible starch coatings on quality aspects of refrigerated Brussels sprouts. *Food Chem* **103**:701–709 (2007).
39 Itoh K, Hatakeyama T, Kimura T, Shimoyama S, Miyazaki A, D'Emanuele A, *et al*. Effect of d-sorbitol on the thermal gelation of methylcellulose formulations for drug delivery. *Chem Pharm Bull* **58**:247–249 (2010).
40 Barreto PLM, Pires ATN and Soldi V, Thermal degradation of edible films based on milk proteins and gelatin in inert atmosphere. *Polym Degrad Stab* **79**:147–152 (2003).
41 Cao N, Yang X and Fu Y, Effects of various plasticizers on mechanical and water vapor barrier properties of gelatin films. *Food Hydrocolloid* **23**:729–735 (2009).
42 Aminlari M, Ramezani R, and Khalili MH, Production of protein-coated low-fat potato chips. *Food Sci Technol Int* **11**:177–181 (2005).
43 Pedreschi F and Moyano P, Oil uptake and texture development in fried potato slices. *J Food Eng* **70**:557–563 (2005).
44 Funami T, Funami M, Tawada T and Nakao Y, Decreasing oil uptake of doughnuts during deep-fat frying using curdlan. *J Food Sci* **64**:883–888 (1999).
45 Katz EE and Labuza TP, Effect of water activity on the sensory crispness and mechanical deformation of snack food products. *J Food Sci* **46**:403–409 (1981).
46 Sánches Silva A, López Hernández J and Paseiro Losada P, Modified atmosphere packaging and temperature effect on potato crisps oxidation during storage. *Anal Chim Acta* **524**:185–189 (2004).
47 Gupta S, Dixit M, Sharma K and Saxena NS, Mechanical study of metallized polyethylene terephthalate (PET) films. *Surf Coat Technol* **204**:661–666 (2009).
48 León PG, Chillo S, Conte A, Gerschenson L, del Nobile MA and Rojas A, Rheological characterization of deacylated/acylated gellan films carrying l-(β)-ascorbic acid. *Food Hydrocolloid* **23**:1660–1669 (2009).
49 Zaccaron C, Oliveira R, Guiotoku M, Pires A and Soldi V, Blends of hydroxypropyl methylcellulose and poly(1-vinylpyrrolidone-co-vinyl acetate): Miscibility and thermal stability. *Polym Degrad Stab* **90**:21–27 (2005).
50 Velazquez G, Herrera-Gómez A and Martín-Polo MO, Identification of bound water through infrared spectroscopy in methylcellulose. *J Food Eng* **59**:79–84 (2003).
51 Filho GR, Assunção RMN, Vieira JG, da S Meireles C, Cerqueira DA, da Silva Barud H, *et al*., Characterization of methylcellulose produced from sugar cane bagasse cellulose: Crystallinity and thermal properties. *Polym Degrad Stab* **92**:205–210 (2007).
52 Passapan W and Chirachanchai S, Conjugation of gallic acid onto chitosan: An approach for green and water-based antioxidant. *Carbohydr Polym* **72**:169–177 (2008).
53 Guillen M D, Ruiz A and Cabo N, Study of the oxidative degradation of farmed salmon lipids by means of Fourier transform infrared spectroscopy. Influence of salting. *J Sci Food Agric* **84**:1528–1534 (2004).
54 Pérez-Mateos M, Montero P and Gómez-Guillén MC, Formulation and stability of biodegradable films made from cod gelatin and sunflower oil blends. *Food Hydrocolloid* **23**:53–61 (2009).

