INFLUENCE OF PROTEIN AND POLYSACCHARIDE BASED COATINGS ON MOISTURE LOSS, FAT UP-TAKE, TEXTURE AND COLOR DEVELOPMENT APPLIED IN COATED POTATO STRIPS DURING DEEP-FAT FRYING.

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ABSTRACT

Under deep-fat frying, food products undergo complex physicochemical changes including mass transfer phenomena (water evaporation and oil migration), generating a crisp exterior crust and moist centre, thereby enhancing palatability and eating satisfaction. Consumers' growing awareness of health issues associated with high-fat foods has increased the demand for low-fat products. Previous studies have shown that hydrocolloid coatings applied to foods prior to frying can act as external barriers and reduce mass transfer. The influence of protein and polysaccharide-based coatings [10% whey protein isolate (WPI), and 1% methylcellulose (MC), respectively], plasticized with either sorbitol (S) or xylitol (X) at concentrations of 0.50%, 0.75%, or 1.00% (w/w), on moisture and oil transfer, color development and mechanical properties was assessed after 60, 120, 180, or 240 s of frying for both non-coated and coated fried potato strips.

The application of WPI coatings plasticized with sorbitol or xylitol increased pre-frying moisture content of raw potato strips by 0.5% (w/w). Percentage moisture loss *(%ML*) of coated samples was significantly lower than that of non-coated potato strips ($P \leq 0.05$). Sorbitol-bearing coatings showed less *%ML* than did xylitol coatings, particularly for shorter frying times (60, 120 s). Across frying times non-coated (*vs.* coated) samples gained significantly ($P \leq 0.05$) greater fat. The most suitable WPI coating formulation in terms of reducing oil up-take was WPI-1.0X (*%FC* = 27.72) at 240 s frying time. The presence or absence of coating had a limited effect on color development. *L*-values decreased (darker) as frying proceeded for both coated and non-coated samples. A substantial increment in coated (*vs*. non-coated) sample texture parameters occurred after 180 s of frying, indicating that much of the desirable mechanical properties were developed at the end of the frying process.

Methylcellulose coatings plasticized with sorbitol or xylitol consistently increased initial moisture content by 0.45% (w/w) of coated potato strips prior to frying. Percent moisture loss *(%ML*) of coated samples was significantly less than that of non-coated potato strips ($P \leq 0.05$), with the lower plasticizer concentration (0.5%) leading to a greater relative decrease in moisture loss particularly during the first minutes of frying (60-120 s). Across all frying times fat content was significantly greater for non-coated than coated

samples ($P \leq 0.05$). The MC-0.5S coating formulation provided the best mass transfer barriers, recording highest values of both moisture content and relative reduction of fat content (MC = 17.83%; *FC* = 36.99%) at last minute of frying (240 s). Coating materials had little to no effect on color development or mechanical properties (F_{max} and Young's modulus). Therefore methylcellulose-based films proved to be an effective film-forming material exhibiting enhanced oil barrier properties compared to the WPI formulations tested.

Mass transfer phenomena (moisture loss and fat content) in both hydrocolloid-coated materials was modeled as a function of frying time using a first order kinetic equation. Moisture loss and fat content rates were greatest early in the frying process. Experimental values fitted well to model-predicted values. The presence of film-forming hydrocolloid (WPI or MC) was strongly tied (higher r^2) to both moisture loss and fat content responses, but not to the nature of the plasticizer (sorbitol or xylitol).

RÉSUMÉ

La friture à l'huile est une opération complexe pendant laquelle les produits alimentaires sont soumis à des changements physico-chimiques comprenant un phénomène de transfert de masse, ainsi que le développement d'une double structure. Combinées, ces propriétés rendent les aliments frits agréables au goût. Cependant, l'accroissement de la conscience sanitaire parmi les consommateurs a augmenté la demande en produits pauvres en matières grasses. Les précédentes recherches ont démontré que des matières hydrocolloïdes peuvent former des revêtements qui agissent comme des barrières externes et réduisent le transfert de masse. Nous avons examiné l'influence des revêtements protéinés composés à 10% de protéines isolées du lactosérum ainsi que celui du polysaccharide composés à 1% de methycellulose. Ces deux matériaux, capables de former des films, sont plastifiés avec du sorbitol et du xylitol dans différentes concentrations.

Les revêtements à base de protéines constitués de protéines isolés du lactosérum plastifiés avec du sorbitol et du xylitol ont démontré leur capacité à augmenter l'humidité initiale présente dans la tranche de pomme de terre avant friture de 0,5%. Pour tous les temps de friture, les échantillons non-recouverts ont gagné significativement ($P \leq 0.05$) les plus grandes valeurs d'assimilation du gras. Les mesures sensorielles ont démontré que la présence ou non d'un revêtement a un effet limité sur le développement de la couleur. Les propriétés mécaniques mesurées montrent une augmentation proportionnelle au temps de friture pour les échantillons couverts ou non.

Les revêtements à base de polysaccharide composés de methylcellulose plastifiés avec du sorbitol et du xylitol montrent une augmentation constante de 0,45% de l'humidité initiale présente dans les tranches de pomme de terre recouvertes avant friture. Le pourcentage de la perte d'humidité dans les échantillons recouverts est significativement inferieur aux tranches de pomme de terre nues ($P \leq 0,05$). Les concentrations les plus basses en plastifiant appliqué (0,50%) conduisent à des taux de perte d'humidité relative supérieurs suggérant que les taux de perte d'humidité sont réduits durant les temps de friture, plus particulièrement lors des premières minutes de cuisson (60-120 s). Pour tous les temps de friture les échantillons nus possèdent significativement ($P \leq 0,05$) les valeurs d'assimilation du gras les plus élevées. En général, la présence des matières de revêtement

démontre une influence marginale sur le développement de la couleur. Les films à base de methylcellulose ont démontré leur efficacité par leur capacité à améliorer les propriétés de barrière contre l'huile, en comparaison aux films à base de protéines isolées du lactosérum.

Des équations mathématiques ont été utilisées dans le but de modéliser le phénomène de transfert de masse pour les deux matériaux hydrocolloïdes testés. Le transfert de masse en tant que temps de friture a été décrit comme une équation cinétique du premier ordre. La perte d'humidité et l'assimilation du gras sont des facteurs significatifs lors des premiers temps de la friture. Les valeurs expérimentales et prédites concordent au modèle proposé. Les matériaux capables de former des films démontrent une forte connexion avec la perte d'humidité et l'assimilation de gras. En revanche, lorsque les données sont regroupées par la nature du plastifiant utilisé, cette connexion est plus faible, donnant des valeurs de coefficient de corrélation inferieures. Cela suggère que la nature du matériel hydrocolloïde utilisé possède un rôle plus important dans le transfert de masse. Les coefficients de corrélation (r^2) les plus élevés sont observés lorsque les données sont groupées par type de plastifiant utilisé pour créer chaque formulation de revêtement.

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CONTRIBUTION AUTHORS

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NOMENCLATURE

ΔE	Equally weighted combination of L, a*, and b* variances
°C	Celsius degrees
a*	Redness-greenness dimensions (CIE L, a*, b* colorspace)
AACC	American Association of Clinical Chemists
ANOVA	Analysis of variance
AOAC	Association of Official Analytical Chemists
b*	Yellowness-blueness dimensions (CIE L, a*, b* colorspace)
C.I.E	Commission Internationale de l'Eclairage
CMC	Carboxymethylcellulose
CRD	Completely randomized design
DM	Dry matter
DMRT	Duncan's Multiple Range Test
FC	Total oil content in control samples
FT	Total oil content in coated samples
GLM	General linear model
h	Hour
HPC	Hydroxypropyl cellulose
HPMC	Hydroxypropyl methylcellulose
HR	Relative humidity
k	Specific rate of reaction
L	Lightness
l	Liter
Mc	Methylcellulose
MC	Moisture content
Mf	Moisture content after frying
min	Minute
ml	Millilitres
mm	Millimetres
Mo	Initial moisture content

N	Newton’s
NSP	Non-starch polysaccharides
O^1_{eq}	Equilibrium oil content
OP	Oxygen permeability
OU	Total oil content reduction
p	p-Value
pH	Hydrogen ion concentration
R^2	Coefficient of determination
s	Seconds
S	Sorbitol
SAS	Statistical analysis software
SPI	Soy protein isolate
SVR	Surface to volume ratio
t	Frying time
w.b	wet basis
w/w	weight of solute over weight of solution
WPC	Whey protein concentrate
WPI	Whey protein isolate
WVP	Water vapour permeability
X	Xylitol

I. GENERAL INTRODUCTION

1.1 Background

One of the most popular cooking methods, deep fat frying allows the elaboration of food products with distinctive quality attributes, including the formation of a dual structure, and the development of distinctive color, flavour and aroma attributes.

The third most important world food crop, potato (*Solanum tuberosum* L.) has seen its production continue to expand under the rising demand of the fast food and snack industry. During the cutting and peeling process irreversible damage of the external cell layers occurs, which can lead to browning and rapid dehydration (Bouchon & Aguilera, 2001). During frying, physicochemical changes in the potato structure take place (*e.g.*, starch gelatinization, protein denaturation, and mass transfer through water vaporization and oil migration, leading to a dual structure: a dry and crisp golden exterior crust and a particularly moist center. A most important textural property found in fried products, crispiness originates from a reduction in moisture content caused by high temperatures transferred from the oil medium (Miranda & Aguilera, 2006).

The first parameter of product acceptance evaluated by consumers is the appearance and color of the food surface (Pedreschi et al., 2005). Color development in fried products is related to Maillard reactions and the reduction of sugars within the potato structure. This occurs at temperatures exceeding 150°C (Miranda & Aguilera, 2006). Fat uptake into fried potatoes largely takes place during the cooling process, after the removal of the samples from the oil medium. Oil located on the surface then migrates into the internal structure due to changes in potato microstructure (Berk, Z. 2009a). Fried potato products' high caloric values arise largely during the deep-fat frying process. Increasing consumer's awareness of the health issues (*i.e.*, cardiovascular diseases) linked to the consumption of fried products has led to the development and production of low oil content fried products. Thus, reducing fat content of fried products has become a major concern for the food industry. The application of edible coatings acting as moisture and fat barriers has proven successful in reducing fat content in fried products (Pinthus et al., 1993; Mallikarjunan et al., 1997; Williams and Mittal 1999; Shaw et al., 2002; Albert and Mittal, 2002; Garcia et al., 2004; Dragich and Krochta, 2010; Tavera-Quiroz et al., 2012). The formulations

studied by the different authors differ in terms of film-forming agent used as well as plasticizer, and concentration applied to create the coating material. In addition, protein and polysaccharides are widely used as film-forming materials, acting as water binders decreasing moisture loss rates, and reducing fat absorption (Pinthus et al., 1993). Water permeability and oxygen permeability are factors that regulate the capacity of a coating to protect a food item. The addition of plasticizers improves flexibility and water vapour permeability resulting in improved films (Shaw et al., 2002). Studies on the influence of coating materials and plasticizer agents on fat content by fried foods and these foods' subsequent quality attributes (*e.g.,* texture, color) could lead to development of enhanced barrier films, further lowering content into such foods.

1.2 Hypothesis of the research

The present work will considerably extend our current understanding of the effect of hydrocolloid materials acting as barriers to mass transfer phenomena and the attendant changes in some quality attributes of coated potato strips. Based on the preceding discussion, the present study's hypothesis is that the application of whey protein isolate (WPI) and methylcellulose (MC) coatings plasticized with Sorbitol (S), Xylitol (X) at different concentrations on potato strips subjected to frying will have significant effect on moisture loss, fat content, as well as color and texture development.

1.3 General Objective

The overall objective of this study was to evaluate the performance of plasticized whey protein suspensions on fat reduction during deep fat frying of potato strips determining how these are influenced by the different concentrations of sorbitol or xylitol, serving as plasticizers. The information could be useful in determining the most suitable formulation for reducing total oil content. Also, a comparison of fat content, moisture loss and overall texture and color development of fried potato strips coated with WPI or MC formulations will be made to determine the best film-forming material. The results were anticipated to enhance quality attributes of coated fried potato strips.

1.4 Specific Objectives

The specific objectives for this work were to:

1. Investigate the effect of WPI coatings containing different plasticizers, [*e.g.*, sorbitol (S) and xylitol (X)], on moisture loss and oil absorption, as well as texture and color development in coated potato strips during deep-fat frying.
2. Study the efficacy of sorbitol and xylitol as plasticizers of methylcellulose coatings on moisture loss, fat content, color development and mechanical properties responses during deep-fat frying of coated potato strips.
3. Determine both mass transfer phenomena (water loss and oil up-take) that take place during frying of coated potato strips described by an empirical first order kinetic reaction.

II. GENERAL LITERATURE REVIEW

2.1 Potato

Potato (Solanum *tuberosum* L.) is produced worldwide and occupies the third rank amongst the world's most important food crops. Native of South America, the white or "Irish" potato was first cultivated by Mesoamericans some 8,000 years ago. Since its introduction into Europe in the 16th century, it has become one of the world's most important food plants, and is now cultivated in over 130 countries (Gould, 1999). A global production of 364 Tg yr^{-1} (2012) demonstrates potatoes to be a leading non-grain commodity in the global food system. China, India, Russia and the United States were the leading producers in 2012 (FAOSTAT, 2012).

Showing a continuous increment in world productions, potato production has particularly expanded in developing countries (Birch et al., 2012) as it produces more food in less time and on less land, than any other major crop. Indeed, the potato has a crucial role to play in reducing world hunger since it offers more calories and protein per acre than any grain crop (Guenthener, 2001). In fact, by 2005 world potato production in developing countries exceeded that of the developed countries for the first time (FAO 2010). The United Nations (UN) declared 2008 to be the International Year of the Potato (IYP), with the mission of increase the awareness of the importance of potato production, highlight the importance of this crop's role in global diets as well as in reducing undernourishment on a global scale (FAO, 2014).

The potato tuber consists of four defined structures: the skin, the cortex, and the outer and inner medullary layers (Brautlecht & Getchell, 1950). The chemical composition of potatoes depends largely on the genetics of each variety as well as the environmental conditions during production and subsequent storage of the crop. Potatoes are rich in starch a carbohydrate that occurs mainly in two forms: amylose (20%) and the balance amylopectin. While carbohydrates constitute roughly ¾ of the total dry matter DM of potatoes, these also provide significant amounts of protein, vitamins, dietary fiber, and are one of the richest sources of antioxidants in human diet. Although potatoes exhibit a high nutritional value, the final total contribution to diet depends largely on the cooking method applied (Storey, 2007). In addition, potato tubers contain phytonutrients including:

polyamines, glycoalkaloids, tocopherols, calystegines, and sesquiterpenes (Navarre et al., 2009).

There are over 200 different wild varieties of Solanum *tuberosum*; however, few of them are of interest for use in industrial prepossess. Selected in 1873 as one of several mutations of the 'Burbank' variety, the 'Russet Burbank' cultivar has become the most popular in North America. The versatility of the potato products depending upon the cooking method applied (*i.e.,* cooked, fried, boiled, roasted, hashed, mashed, etc.) (Gould, 1999b).

The growth in potato production is also tied to an increased demand from the fast food industry for frozen and fried potato products. Fried potato products compromise a large proportion of the total potato crop production; 30% is destined to frozen French fries and 12% towards chips and shoestring products (Miranda & Aguilera, 2006).

2.2 Deep-Fat Frying

Potato is a high caloric food in itself, and frying further increases its caloric content. Understanding frying is of great interest to commercial potato processors, especially the food service and snack industry, where potato is served in the form of French fries and chips (Odenigbo et al., 2012). Deep-fat frying is defined as a process by which a food item is cooked by the total immersion of a food item in an oil medium (Ngadi et al., 1997). Occurring in oil heated to between 160°C and 180 °C, frying is a complex cooking and drying process in which heat and mass transfer occur simultaneously. Convective heat is transferred from the oil to the food surface, and then conductive heat transfer into the food core. In addition, mass transfer occurs due to realise of water and oil absorption. A moving boundary is produced within the food separating the formation of the external crust from the core that is being cooked (Miranda & Aguilera, 2006). Orthoefer (2006) defined heat transfer under frying as the movement of energy from the heat source to the oil and finally to the food item being fried. While temperature differences determine the rate of heat transfer, the thermal properties of the oil and food item are key factors in designing an optimal frying process.

The overall quality of a fried product can be determined by measuring the related product properties, which include: geometric properties (size, shape surface area, volume

and density), thermal properties (thermal conductivity, thermal diffusivity, specific heat, heat transfer coefficient), mass transfer properties (moisture diffusivity, fat diffusivity, mass transfer coefficient), mechanical properties (hardness, cohesiveness, viscosity, textural properties), and optical properties (Mittal, 2008). Similarly, Pedreschi and Zúñiga (2009) stated that the heat and mass transfer during frying can be affected by food properties, oil composition, and food geometry and oil temperature. Heat transfer from the oil medium to the product can also be retarded due to the formation of an insulating layer of vapour on the surface of the potato.

2.3 Physical and chemical changes in fried potatoes

During deep fat frying food products undergo complex physicochemical changes such as protein denaturation, starch gelatinization, water vaporization and crust formation; all together these make fried foods palatable and provide special eating satisfaction to the consumers (Porta et al., 2012). Significant microstructural changes in fried products are also observed; in potatoes, crust formation is the result of several alterations, most of them occurring at the cellular and subcellular level. These changes mostly occur in the external cell layers, which suffer physical damage due to the peeling and cutting process, gelatinization of starch granules, softening of cell walls, and rapid dehydration. The major changes are hydration and swelling of starch granules and softening of the middle lamellae. In contrast, changes observed in the core are much milder (Bouchon & Aguilera, 2001). According to Ngadi al. (2010) the development of pores is a major structural change during frying, caused by intense heat that could lead to explosive evaporation resulting in the formation of pores and crevasses. These pores differ in size, shape and distribution, which significantly affect the final quality attributes of the fried potato (i.e. mechanical, sensory and textural properties).

During frying, a dual structure is formed: a dry and crisp golden crust and a moist center. This dual structure, in part, occurs because there is a reduction of water content from 75-85% to 1-2% w.b causing a partial shrinkage (65%) of the material (Miranda et al., 2006). Moreover, O'Connor (2001) studied the responses on moisture loss of different potato varieties during deep-fat frying concluding that moisture loss rates differed between cultivars due to a differences in total solids, sugar and starch content, specific gravity, and

different physicochemical properties. Gamble and Rice (1987) found the crust to be formed during the most deep fat frying processes as having a direct influence on heat and mass transfer phenomena, oil up-take and physical properties of fried products.

Debnath et al. (2009) studying the differences in oil absorption of surface vs. internal structures of fried potato slices, found that there are two main oil fractions: surface oil and structural oil (oil content in the core). After frying, oil located in the surface migrates into the structure due to condensation of vapour inside the product creating a vacuum. Berk, (2009a) noted that the system - oil medium and potato - maintains an equilibrium between the moisture loss, water evaporation, and the fat transfer that occurs during frying due to changes in the microstructure. Kinetic reactions are also involved due to the loss of moisture content and the relation between the presence or absence of a crust in the external surface of the product. Berk (2009b) found that after deep fat frying potatoes strips showing proper attributes exhibited a dual structure, a dry and crispness texture, a golden crust and a particularly moist center.

Studying kinetics of oil into fried potato slices, Duran et al. (2007) determined that the full oil content of the samples was absorbed during the first 30 s of frying, where most of the oil was located principally in the surface of the slice, and that after its removal from the frying medium, the oil tended to penetrate into the product's microstructure.

Since a greater part of the oil is absorbed after removal from the oil medium, the conditions under which potatoes strips are removed from the fryer are important in regulating oil absorption. After it is cooled, the greatest quantity of oil is retained in the crust of the fried material in the form of droplets (Pedreschi et al., 2008). Similarly, Moreira et al. (1997) found that in fried tortilla chips about 64% of oil up-take takes place during the cooling period, due to a rise in internal tension between oil and air as the temperature decreases, resulting in a rapid passage of oil from the external surface into the porous structure of the chip.

Formation of acrylamide compounds are also linked to frying process. According to Matthaus et al. (2004) the presence of low molecular protein components (i.e. aminoacids) and a reduction in sugar (fructose and glucose), along with water loss and high temperatures are relevant factors in acrylamide formation. Kita et al. (2004) stated the formation of acrylamide is highly dependent on temperature and time of frying; suggesting

that to decrease the formation of acrylamide compounds oil temperatures should not exceed 175°C and frying time should be shortened. However, lower temperatures during frying will influence product fat content and moisture loss. Moisture loss is one of the critical quality factors because it affects the texture of the final product.

Taubert et al. (2004) noted that the effect of temperature on the formation of acrylamide depends on the surface-to-volume-ratio (SVR) of the food product. Later, Granda and Moreira (2005) established that potato products with low SVR exhibited greater acrylamide formation.

2.3.1 Mechanical Properties

During deep-fat frying, the structure of raw products undergo major changes, which are related to the overall quality of the final product therefore, these ones influence in consumer's acceptance of the food product. Texture is a multi-parameter attribute associated with mechanical, geometrical and acoustic parameters (Szczesniak, 2002). Crispness is a major textural property of fried products resulting from the reduction in moisture content caused by high temperatures applied during frying (Miranda & Aguilera, 2006).

Although flexural and impact strength, and elasticity modulus of fried potatoes tend to increase over frying time (Pinthus et al., 1995), the force-deformation curve is not consistent due to irregular shapes, sizes and curvatures in the samples (Segnini et al., 1999). According to Aguilera and Gloria (2000) the development of crispy structure starts after the first minute of frying. The crispy surface originates in the migration of oil to intracellular spaces formed during frying. However, crust formation in fried potatoes does not lead to the rupture of cells, which remain intact but shrunken and dehydrated with swollen starch granules. Lisińska and Golubowska (2005) described structural changes of potato tissue during French fry production, and textural changes in French fries were due to water loss, irreversible damage of potato tissue and changes in non-starch polysaccharides (NSP) and lignin contents.

A kinetic approach of textural development in potato tissue done by Pedreschi et al. (2001) showed that the initial stages of frying lead to tissue softening, cooking the core and the onset of crust formation. This resulted in a product exhibiting a composite structure

made of a hard crust region and a soft-core interior. They further demonstrated that starch content was a determining factor in textural development of fried potatoes, causing a rounding of cells and their separation, whereas pectic materials prevent cell separation and contributed to cohesiveness. Similarly, Kita (2002), studying the influence of chemical composition on crisp texture in five different varieties of potatoes, found texture development to depend on the content of starch, nitrogen substances and non-starch polysaccharides (*e.g.*, protopectins), to have the most important influence on textural development.

2.3.2 Color development

Krokida et al. (2001) defined color as the sensation experienced by a person when energy in the form of radiation within the visible spectrum falls upon the retina of the human eye. The appearance and color of the food surface in a food product is the first quality parameter evaluated by consumers being critical in the acceptance of the final product (Pedreschi et al., 2005b). Kudra and Strumillo (1998) stated that changes in colors of fried products are due to evaporation of surface water and oil uptake, as well as, enzymatic browning, non-enzymatic browning and caramelization reactions. Color changes in fried potatoes occur mainly because of Maillard reactions between amino acids or free amino groups of protein peptides, and reducing sugars, mostly glucose and fructose. Browning in potatoes strips increases at oil temperature $T_{oil} \geq 150°C$ (Miranda & Aguilera, 2006). Marquez and Añon (1986) also noted that the color of fried potatoes is the result of Maillard reactions, whereby brown pigments are formed in oil heated potato slices through a reaction between sugars and amino acids. In such reactions, the amount of reducing sugars becomes the limiting factor. Fructose has been demonstrated to generate the strongest browning reactions, followed by glucose; surprisingly, the addition of the disaccharide sucrose had no significant effect on the final color of fried potatoes.

Pedreschi et al. (2005b) determined color development only begins when sufficient amount of drying has occurred in potato slices and that this is dependent on drying rates and the overall heat transfer coefficient during the first stages of frying. Moreover, they defined color development as a surface phenomenon where coloring rates are linked to

potato slices, thickness, moisture loss rates and the temperature of the oil medium during frying.

Krokida et al. (2001) stated that potato lightness (L) increased during the early stages of frying, but then remained constant as frying progressed. As other, they found that oil temperature, type of oil, frying conditions, sample thickness, and type of pre-treatment applied were all factors, which affected color development kinetics.

Tajner-Czopek et al. (2008) studying the effects of potato strip size and pre-drying method on French fry color development showed that pre-drying of potato strips led to an increase in optical properties based on changes in the intensity of green (↓ a* values) and yellow (↑ b* values) colors, as well as, higher lightness (L) values compared to non-pre-dried French fries.

The appearance and color of the food surface in a food product is the first quality parameter evaluated by consumers being critical in the acceptance of the final product (Pedreschi et al., 2005b).

2.4 Methods applied to reduce fat absorption.

In order to reduce oil up-take during frying and produce relatively low fat products, it is necessary to study and understand all the mechanisms involved during the frying process. Many factors have been reported to affect oil content into French fries, including oil quality, frying time and temperature, and content (moisture content, solids, fat, gel-strength, and proteins), pre frying techniques (i.e. drying, blanching, frying) and the presence of coating materials (Pinthus et al., 1995). Gould (1999c) pointed out that a careful selection of varieties with high specific gravity, thick slices with less potato surface area, and a high frying temperature for a shorter time can reduce oil absorption. Moreover, modification of frying techniques, including the proper shaking and draining of the fried food after removal from oil frying medium can remove excess surface oil content (Mellema et al., 2003). Garayo and Moreira (2002) demonstrated that vacuum frying has is an effective method to produce chips of lower oil content, which nonetheless exhibit appropriate color development and textural properties. Aguilera et al. (1997), suggested that blanching at lower temperatures could contribute to the reduction of fat absorption by French fries. Similarly, Al-Khusaibi et al. (2012), found that blanching at 65 °C for 5

minutes was enough to cause a total gelatinization of potato starch which might result in less oil content in the final product. Many factors have been reported to affect oil absorption in fried foods. These include oil quality and composition, frying time and temperature, product composition, moisture content, surface treatments, initial interfacial tension, and crust size (Gamble & Rice, 1987; Pinthus & Saguy, 1994).

2.4.1 Influence of oil quality on fat absorption.

During deep-fat frying, the fat is exposed continuously, and repeatedly to elevated temperatures in the presence of air and moisture. In addition, chemical reactions take place, including oxidation, hydrolysis, and polymerization producing volatile and non-volatile compounds (Stevenson et al., 1984). According to Choe and Min (2007) most of the volatile compounds evaporate in the atmosphere with steam and the remaining volatile undergo further chemical reactions or these ones are absorbed in fried foods. Non-volatile compounds in the oil are responsible to modify physical and chemical properties of the oil medium and fried products; affecting flavor stability, overall quality and texture of fried foods during storage (Choe & Min, 2007).

Frying oil quality have certain influence on oil absorption due to the formation of oil degradation compounds (high molecular weight polar compounds) which increase polarity of the frying medium. Consequently, oil viscosity increase which increases the amount of oil on the food surface (Rimac-Brnčić et al., 2004). Food fried in oil with high viscosity tends to absorb more frying oil resulting in foods with soggy, greasy, and less appetizing aspect (Orthoefer et al., 2006). Kita et al. (2005) reported that there is a significant correlation between fat content of French fries and the content of saturated, unsaturated, essential and trans isomers fatty acids of frying medium. Fat absorption increased with increasing unsaturated fatty acids and decreasing saturated fatty acids, essential fatty acids and trans isomers fatty acid content. The level of absorbed fat by fried products increases with increasing oil degradation (Dobarganes et al., 2000).

Therefore, frequent replenishment of fresh oil has been reported in order to decrease the formation of polar compounds, diacyglycerols, and free fatty acids resulting in an improved frying life and quality of oils (Romero et al., 1998).

2.4.2 Influence of frying time and temperature on fat absorption.

During frying process the time and temperature have been reported to have a close relation to fat absorption rates on fried products. Oil penetration can only occur where temperature has been sufficiently high to cause water evaporation, pore formation, i.e. in the crust (Mellema, 2003). Gamble et al. (2007), a lower oil temperature resulted in a lower oil content in early stages of frying with greater differences between 145 °C and 165 °C than between 165 °C and 185 °C. In general, longer frying time leads to more water evaporation, thus more external damage and pore formation resulting on higher fat absorption. Oil absorption increased until the frying time of 180 seconds when this rate is diminished (Gamble et al., 2007). Moreover, high temperatures of oil (180 °C) allows rapid heat transfer and short cooking times resulting in less final fat content.

2.4.3 Influence of food composition and initial moisture content on fat absorption.

The water present in food prior frying play a crucial role as it gets converted into steam during frying process. This steam cooks the food item and then escapes through pores due to internal pressure. The voids left by the removal of water through pores are then filled with fat, especially in the outer layer (Mehta et al., 2001). Therefore, initial moisture content significantly affect the final oil content during frying, as initial moisture content increased the final oil content increased on fried tortilla chips (Moreira et al., 1997). Similarly, Gamble and Rice (1988), concluded that initial and final water content of fried chips are closely related to total fat absorption during deep-fat frying. In addition, Gamble et al. (1987) reported a linear relationship between total fat content and water evaporation. Sayre et al. (1990) fat absorption depends more on the surface moisture content than on the total moisture content of fried chips.

Makinson et al. (1987) reported that plan foods, which initial high water and low fat contents, absorb more frying fat than animal foods. In addition, Irmiter et al. (1967) stated the presence of high levels of fat in raw meat retards water evaporation. Animal tissues are filled with fluids, those of plant tissues are filled with air, thus plant foods result in greater capacity to retain absorbed fat (Fillion et al., 1998).

Specific gravity and solids-content have also been related to have an effect on fat absorption in fried potato strips. Potatoes with high specific gravity recorded to produce

high yield of French fries with a lower fat content (Lulai & Orr, 1979). In addition, potatoes with higher dry mass content produce crisps with lower fat content than those with lower dry matter values (Kita, 2002).

2.5 Coatings and Edible films

An edible film is defined as a thin layer, placed as barrier between the food and the surrounding environment, which may or may not be consumed. Because edible films act as barriers, reducing mass transfer phenomena (i.e. moisture, flavours, and gases), they can serve to reduce microbial contamination, which can, in turn, lead to quality losses in processed foods. Edible films can also be used as antimicrobial agents, antioxidants, enzymes or functional ingredients such as probiotics, or sources of minerals or vitamins. Edible films can be classified depending on their nature of their components: hydrocolloids (containing proteins, polysaccharides or alginates), lipids (formed based on fatty acids, waxes), and composites (combinations of the two previous types) (Skurtys et al., 2010). Capable of exhibiting hydrophilic or hydrophobic properties, coatings are composed of natural film-forming materials such as hydrocolloids, proteins, lipids or combinations of these three (Zaritzky, 2011).

Bearing many hydroxyl groups hydrocolloids are hydrophilic polymers, obtained from animal, microbial, vegetable or synthetic source. Currently, hydrocolloidal materials (*i.e.*, protein and polysaccharides) are widely used as film-forming agents, to increase viscosity in an aqueous phase, and, based on this ability, as emulsifiers, due to their emulsion-stabilizing properties (Skurtys et al., 2010). According to Pinthus et al. (1993) hydrophilic polymers in a coating can be used as water binders to reduce water loss, resulting in the case of French fries, in a reduction in oil content.

At the moment a coated food product is fried, the film hinders oil absorption, resulting in a reduction of fat absorption and calories, and, an improvement in nutritional qualities of the final product (Mallikarjunan & Phillips, 1997). Several authors have studied the properties of different coatings to reduce oil absorption (Mackinson et al., 1987; Debeaufort & Voilley, 1997; Huse et al., 1998; Williams & Mittal, 1999; Garcia et al., 2004; Suarez et al., 2008).

Coatings enhance the quality of food products protecting them from physical, chemical and biological deterioration, acting as barriers against oils, gases, or vapours. Coatings exhibiting high hydrophilic properties mainly act as sacrificing agents, as in the case of high-moisture gelatines acting as barriers to moisture. However, they can also be used to prevent lipid oxidation due to them providing good oxygen barriers (Han & Gennadios, 2005; Kester & Fennema, 1986). Garcia et al. (2004) stated that the effectiveness of a coating is determined mostly by its mechanical and barrier properties, which are related to its composition and microstructure, as well as other characteristics of the substrate to which it is applied. According to McHugh et al. (1994) coatings regulate the transfer of moisture, oxygen, carbon dioxide, lipids, as well as aroma and flavour compounds in the food systems.

Film-forming solvent systems and conditions during film formation determine the final characteristic of the film. Cohesive strength and flexibility are critical factors that define porosity, permeability and uniformity of thickness of the barrier and its adhesion to the coated product. Furthermore, the thickness of the coating plays an important role on the efficacy of the film to act as an oil and moisture barrier (Varela & Fiszman, 2011). Similarly, Tavera-Quiroz et al. (2012) indicated that good film adhesion and flexibility are critical factors in integrity of each coating formulation as they decrease the likelihood of discontinuities and brittle zones. Moreno et al. (2010) indicated that a food's surface roughness, along with other physical properties, are key factors in a fried food's response to a given coating material, and associated oil content. Baldwin et al. (2012) noted that water vapour permeability (WVP) and oxygen permeability (OP) were the barrier properties that usually determine the capacity of a coating to protect the food product from the environment. The film's WVP, OP, and mechanical properties depend upon its composition and structure.

The application of edible coatings for reducing oil content of fried foods can be used to produce products that meet both the health and quality preferences of the consumers (Garmakhany et al., 2008).

2.5.1 Protein based coatings

Proteinaceous film-forming materials are derived from different plant and animal sources. Milk, whey, casein, collagen (animal) and soybean [Glycine max (L.) Merr.], wheat [*Triticum aestivum* L.], and corn [*Zea mays* L.] proteins (*e.g*., zein), are often used as materials to produce films. Protein suspensions provide better oxygen and carbon dioxide barriers and mechanical properties than polysaccharide coatings, but present poorer water barrier properties. Protein films are very brittle without addition of plasticizers; however, their addition affects the properties in protein-based films. Plasticizers are needed to provide flexibility to protein-based coatings, and to increase water vapour permeability. The solubility of the film in water depends upon the protein source used and the conditions of the film formation and treatment (Skurtis et al., 2010).

Whey proteins remain in milk serum after cheese manufacture, and represent about 20% of total milk proteins (Brunner, 1977). Depending on their protein content they can be classified as: whey protein concentrate (WPC; 20-80%) or whey protein isolate (WPI; >90%). Whey protein films are transparent, bland and flexible, with excellent oxygen, aroma and oil barrier properties, but only provide a poor moisture barrier due to their hydrophilic properties. Whey proteins are globular and heat labile in nature, consisting of several component proteins, including α-Lactalbumin (α-La), β-Lactoglobulin (β-Lg), bovine serum albumin (BSA), immunoglobulins (Ig), and proteosepeptones (PP) (Kinsella & Whitehead, 1989).

Table 2-1 Protein and mineral composition of whey protein isolated.

Component	Microfiltration WPI	Ion exchange WPI
α - Lactalbumin	15-22	14-26
β - Lactoglobulin	56-60	66-75
Bovine serum albumin	1 – 2	3 – 6
Immunoglobulins	2 – 5	2 – 3
Glycomacropeptides	20-26	Not detected
Lactoferrin	0-0.1	Not detected
Peptide fragments	3 – 5	Not detected
Calcium	0.3-0.6	0.08-0.11
Sodium	0.2-0.3	0-0.5

Huffman & Barros Ferreira (2011).

When heat treatment is included in film formation, a denaturation of the protein can generate water insoluble films with enhanced mechanical properties. Film formation under these conditions can be influenced by the protein's amino acid composition, as well as their distribution and polarity, factors related to the formation of ionic crosslinks between amino and carboxyl groups, hydrogen bonding groups and intra and intermolecular S-S bonds (Gennadios & Weller, 1991). Crosslinking reactions are pH dependent, decreasing as pH increases over 7.5 (Dangaran & Krochta, 2009). Heat denaturation above 65 ◦C, opens the β-Lg globular structure, exposes sulfhydryl and hydrophobic groups, and induces oxidation of free sulfhydryl, disulfide bond interchange, and hydrophobic bonding. These reactions can be used to form water-insoluble edible films (Khwaldia et al., 2004).

Coatings derived from WPI require the addition of plasticizers to overcome brittleness; however, such addition can cause significant shifts in WPV and mechanical properties of the films. The most common plasticizers in WPI suspensions are glycerol, sorbitol and polyethylene glycol (Baldwin et al., 2012).

A whey protein film network can also be affected by changing the free volume of the matrix by the addition of an internal or external plasticizer. Internal plasticizers are molecules, which can chemically modify a chain (*i.e.*, acylation, carboxylation), whereas external plasticizers act as lubricant in WPI films by interrupting protein-protein interactions, thus allowing greater movement is possible. Plasticizers have to be compatible in size with the polymer, shape, and chemistry to be effective. Plasticizers act by disrupting hydrogen bonding between polymer chains thus increasing chain mobility and decreasing brittleness (Kester & Fennema, 1986).

McHugh et al. (1994) heat treatments allowed the formation of intermolecular disulphide bonds resulting in water-insoluble whey-protein-based films. Optimal conditions for preparing WPI films included heating of 10% (w/w) WPI solutions at 90°C for 30 min. The report included the study of the influence of sorbitol, glycerol and polyethylene glycol as plasticizers in WPI films. While they showed that plasticizers improved film flexibility, they also showed that, depending on the particular plasticizer, this led to different changes in film permeability. Films plasticized with sorbitol exhibited higher relative humidity (HR) and lower values of WVP, than the values properties exhibited by glycerol plasticized films.

Similarly, Shaw et al. (2002), studying the effects of glycerol, xylitol and sorbitol on physical properties of WPI films, concluded that increasing glycerol or sorbitol concentrations led to an increase in moisture content, WVP, along with a decrease in tensile strength, elastic modulus, and glass transition temperatures of the films. In addition, an increment in film flexibility was observed on higher concentrations of plasticizer added. Results suggested that plasticizers might function by changing moisture content of the films.

Perez-Gago and Krochta (2002) stated that native and heat-denatured WPI films have similar WVP; however, they exhibit different solubility's and mechanical properties. Plasticized WPI films are poor moisture barriers, but their permeability can be reduced by the incorporation of lipid materials at temperatures above their melting points into the aqueous film-forming suspensions of heat-denatured plasticized WPI films. All WPI suspensions, with or without lipid emulsification, must be degassed until no more air bubbles are observed, to ensure an accurate assessment of permeability values.

Dragich and Krochta (2010) investigating the effect of WPI coatings on the reduction of oil content in fried chicken found no significant differences between coated and uncoated chicken strips in terms of fat up-take.

2.5.2 Cellulose derivatives coatings

Cellulose suspensions are polysaccharides materials composed of linear chains with the presence of methyl, hydroxypropyl or carboxyl substituents. Methylcellulose (MC), Carboxymethylcellulose (CMC), hydroxypropyl cellulose (HPC) and hydroxypropyl methylcellulose (HPMC) are the most common polysaccharides materials used to form coatings. A physical hydrogel may exhibit reversibility that the gel can change its physical state from a liquid (solution) to a solid (gel), and vice versa, corresponding accordingly to the external stimuli such as temperature, pH, electric field, magnetic field, salt, surfactant, solvent composition and light (Li, 2002). In aqueous solutions, the gelation from hydrophobically modified cellulose is considered to be due the inter-molecular association of the hydrophobic groups on the polymer chains, which is a function of temperature (Li, 2002). In addition, water molecules when subjected to low temperatures form "cagelike" structures to surround the hydrophobic methoxyl groups, causing the MC

to become water-soluble (Sakar & Walker, 1995). Once it is subjected to heat treatments, these structures distort and break to expose the hydrophobic regions, inducing the formation of aggregates, thus gels are formed (Kobayashi et al., 1999). Methylcellulose films form gels at high temperatures, where hydrophobic polymer chain interactions are involved in the thermal gelation process (Zaritzky, 2011). Cellulose suspensions exhibit thermo-gelation properties, *i.e*., when are exposed to high temperatures they form gels, but when cooled the film returns to the originally consistency. Hydrophobically modified cellulose exhibit a unique property of disruption of the associative aggregates or junctions formed at elevated temperatures causing a return to liquid state again upon cooling through the disassociation of hydrophobic groups, property known as "thermoreversibility" (Li et al., 2002).

Cellulose based coatings are resistant to oil and fats, show moderate strength, are flexible, transparent, colorless, tasteless, and water-soluble and provide a moderate barrier to oxygen (Skutys et al., 2010). Pinthus et al. (1993) observed that the increase in firmness due to thermogelling resulting in gels with a higher strength, which results in a lesser moisture loss and lower water diffusivity.

Thermogelling properties in MC coatings encourage cohesion and adherence (film pick-up) to the surface of the substrate. Viscosity of the coating formulations is a rheological property, which has an important role in conferring proper adherence of to the material (Tavera-Quiroz et al., 2012). Williams and Mittal (1999) found that MC coatings showed the best barrier properties in terms of reducing of fat absorption into the several food products tested (frozen potatoes, rehydrated potato starch, mashed potatoes and commercial pastry mixes).

Albert and Mittal (2002) studied eleven different hydrocolloid materials including polysaccharide and protein-based coatings on water and fat transfer properties. They demonstrated that soy protein isolated (SPI), whey protein isolated (WPI), and methylcellulose (MC) exhibited the best performance on fat reduction during frying of a cereal product.

Garcia et al. (2004) investigating the influence of the addition of different concentrations of sorbitol as plasticizer in MC solutions, found that MC suspensions without plasticizer showed cracks (fractures) in the coating, whereas the presence of

sorbitol improved coating integrity, resulting in films providing enhanced oil and moisture barriers. The most effective formulations was 1% MC with 0.5% sorbitol for potato strips exhibiting a 40.6% of oil reduction.

CONNECTING TEXT

Overall quality in fried products has been reported to be affected by several factors such as oil quality, frying time and temperature, physical and chemical properties of food items, application of pre-frying techniques, and the presence of coatings materials. The use of proteins as ingredients in fried products is also known, yet their functionality as coatings to improve quality in fried products have not been established. In chapter III, the influence of whey protein isolated based coatings on quality of coated potato strips during deep-fat frying was assessed. Effects on moisture loss, fat absorption, color development and mechanical properties were evaluated.

III. INFLUENCE OF WHEY PROTEIN ISOLATED-BASED COATINGS ON QUALITY OF POTATO STRIPS DURING DEEP-FAT FRYING.

3.1 Abstract

Reducing oil content in fried products by the application of coatings allows the production of healthy foods, which meet consumers' preferences. A protein-based coatings composed of whey protein isolate (WPI), plasticized with sorbitol (S) or xylitol (X) at concentrations of 0.50, 0.75 or 1.00, were applied to potato strips prior to frying. Oil and moisture transfer, color development and changes in textural properties after 1, 2, 3, or 4 min of frying were assessed in non-coated (control) and coated (WPI) fried potato strips. Initial moisture content in coated potato strips exhibited an increment of 0.5% relative to non-coated samples prior frying due to the presence of the coating film. Film pick-up rates showed no significant differences between sorbitol and xylitol coating formulations ($P > 0.05$). Potato strips coated with WP-xylitol 1.00% formulation exhibited highest initial moisture content (83.32). Xylitol-coating formulations exhibited higher moisture content (MC) in average than did sorbitol coatings; however, plasticizer concentration added in coating formulation had no significant effect ($P > 0.05$). Across all frying times non-coated samples gained significantly ($P \leq 0.05$) greater fat content values (FC = 20.07%) than any of the coated slices. The most suitable WPI coating formulation regarding the relative reduction of oil up-take in percentage was WP-1.0X (27.72) at 240 s frying time. Regarding color development L-values decreased (darker) as frying proceeded for both coated and non-coated samples. The presence or absence of the coating material or the nature of the plasticizer added had relatively little influence on color development. Mechanical properties increased over frying time for both coted and non-coated samples. A substantial increment in texture parameters relative to non-coated samples were recorded after 180 s of frying, indicating that much of the desirable mechanical properties were developed at the end of the frying process. Moreover, force at maximum load (F_{max}) as a function of hardness and Young's modulus (λ) as a function of stiffness in coated potato strips were greater than in non-coated samples; however, these differences were not significant ($P > 0.05$) with any treatment factor with the exception of frying time. Frying time had significant effects ($P \leq 0.05$) on all variables relative to non-coated samples analyzed (MC, $\%FC$, L, a, b, ΔE, F_{max}, and λ).

3.2 Introduction

Deep fat frying consists of cooking a product in hot oil. The oil's high temperature results in the product losing water by evaporation, and gaining oil to replace the lost water. During frying, unique physical, chemical, and sensorial attributes are acquired. Texture, color, and oil content are among the main parameters of interest in fried products (Basuny et al., 2009).

The quality of fried potatoes depends largely on their structural, textural, and optical properties (Moreira et al., 1995; Farkas et al., 1991). Texture is a multi-parameter attribute associated with mechanical, geometrical and acoustic parameters (Szczesniak, 1987). When products of relatively high starch content (*i.e.*, potatoes) are heated, their textural properties change as a result of starch gelatinization (Pedreschi & Moyano, 2005). The flexural and impact strength and elasticity modulus of fried potatoes tend to increase over frying time (Pinthus et al., 1995). Color changes in fried potatoes occur because of Maillard reactions between amino acids or free amino groups of protein peptides, and reducing sugars, primarily glucose and fructose. Browning rates in fried potatoes strips increases when oil temperatures exceeds 150°C (Miranda & Aguilera, 2006).

Crispiness as a major textural property of fried products results from the reduction in moisture content caused by high temperatures applied during frying (Miranda & Aguilera, 2006). Fried products show a crispy crust and a soft moist interior, which increases their palatability (Mallikarjunan et al., 1997). However, given the public's increasing awareness of the need for reducing the proportion of fat in the average diet, has prompted study into ways to reducing oil content in fried foods (Gamble et al., 1987). The application of edible coatings designed to reduce oil content of fried foods can result in the production of fried foods that meet both the health and quality preferences of the consumers (Garmakhany et al., 2008). Coatings are able not only to extend the product's shelf life, but also to improve its quality as well. Coatings regulate the transfer of moisture, oxygen, carbon dioxide, lipids, aroma, and flavour compounds in the food systems (McHugh et al., 1994). A coating's effectiveness is mainly determined by its mechanical and barrier properties. These are related to its composition and microstructure (Garcia et al., 2004). Adhesion and flexibility are critical factors in the integrity of each coating formulation as they contribute in decreasing possible discontinuities and brittle zones (Tavera-Quiroz et al., 2012).

Several materials can be used in formulating coatings. Possible materials include gelatin, gellan gum, methylcellulose (MC), microcrystalline cellulose, pectin, sodium caseinate, soy protein isolate (SPI), wheat gluten and whey protein isolate (WPI).

Protein coatings composite by 10.68% WPI and 9% SPI have provided far greater fat-uptake reduction than polysaccharides such as methylcellulose in dough products (86%, 80% and 58%, reduction, respectively) (Albert & Mittal, 2002).

Whey protein coatings form transparent, bland, and flexible films, excellent in controlling oxygen, flavour, aroma, and oil transfer between food components and the atmosphere surrounding the food item (Baldwin et al., 2012). However, in the absence of plasticizers, whey protein films are very brittle. Therefore, to provide flexibility to whey protein-based coatings the addition of plasticizers is required although such an addition may increase water vapour permeability (Skurtis et al., 2010). Plasticizers reduce internal hydrogen bonding and increase intermolecular spacing, causing reduction of brittleness, which translates to an increase in permeability of the film materials (Lieberman & Gilbert, 1973). Heat treatment is necessary to allow formation of intermolecular disulphide bonds by thiol-disulfide interchange and thiol oxidation reactions (McHugh et al., 1994).

Ozdemir and Floros (2008) studied the optimization of edible whey protein films containing different concentrations of sorbitol, beeswax, or potassium sorbate, in order to improve mechanical and optical properties of food products. In addition, Shaw et al. (2002) demonstrated that the proportional increase in film flexibility achieved by increasing the quantity of plasticizer was the result of a decreasing glass transition temperature and increasing equilibrium moisture content of the films. Plasticizers may function by altering moisture content of films (Shaw et al., 2002). Glycerol, sorbitol, and polyethylene glycol have been commonly applied in whey protein coating formulations (Perez-Gago & Krochta, 2002). However, most of the studies composite films have been devoted to the characterization of the mechanical properties and permeability of edible films (Khwaldia et al., 2004). Less attention has been paid to the application of them in order to determine their effectiveness acting as barriers properties reducing mass transfer phenomena during deep fat frying.

Albert and Mittal (2002) studied the effect of eleven different film materials on water and fat transfer properties in a pastry mix including isolated whey protein. Results obtained demonstrated that WPI formulations were able decrease water evaporation by 35.97% and to reduce 86% total fat absorption. However, previous work describing protein coatings for fat absorption reduction in fried chicken have reported limited success. In 2006, Ballard and Mallikarjunan recorded no differences in fat content between applications (WPI and MC added to predust compared with WPI and MC added to batter). Dragich and Krochta (2009) investigated the application use of denatured isolated whey protein (DWPI) as an additional

coating to chicken breast previously coated with wheat flour – batter- wheat flour (WF-batter-WF). Results revealed that chicken breast coated with WF-batter-WF dipped and then dipped in 10% DWPI solutions before frying recorded a reduction of 30.67% of fat content. Dogan et al. (2005) incorporated 3% SPI, 3% WPI or 3% egg albumin (EA) into a batter applied to chicken nuggets. Results recorded no significant differences between each other in terms of fat absorption, and both EA and WPI had significantly less fat (11% & 13%, respectively) than both SPI and control samples (no protein added). The fat reduction for WPI treatments was much less than the 86% fat reduction recorded by Albert and Mittal (2002) on dough balls.

Previous research has shown inconsistent results regarding the effectiveness of WPI coatings as barriers for fat reduction in fried products this could be attributed to the differences in concentration of protein applied, film formation from heat denatured protein prior to application in the food item, and the incorporation of WPI as an external layer rather than incorporated to the product itself (Dragich &Krochta, 2009).

The objective of this research was to investigate the effect of WPI coatings containing different plasticizers, sorbitol and xylitol, on moisture content, oil absorption, texture and color attributes of coated potato strips during deep-fat frying.

3.3 Materials and methods

3.3.1 Protein based suspensions

Coating ingredients included whey protein isolate (Canada Protein, ON, Canada (29 g protein per 30 g serving), sorbitol (Fisher Scientific, ON, Canada) and xylitol (Fisher Scientific, ON, Canada). Aqueous suspensions of 10% whey protein isolate (WPI) were used, as films did not form below 8% whereas gels were formed above 10% (McHugh et al., 1994). Sorbitol (S) and xylitol (X) plasticizer concentrations of 0.50, 0.75, and 1.00% (w/w) were tested, resulting in six coating formulations: WPI-sorbitol 0.5% (WPI-0.5S), WPI-sorbitol 0.75% (WPI-0.75S), WPI-sorbitol 1.0% (WPI-1.0S), WPI-xylitol 0.5% (WPI-0.5X), WPI-xylitol 0.75% (WPI-0.75X) and WPI-xylitol 1.0% (WPI-1.0X). The WPI (15 g) was weighed and slowly dispersed in 150 ml of distilled water under constant stirring. Solutions were heated up to 90 ± 2°C for 30 min. allowing film formation to occur. Plasticizers were incorporated into the solution after the WPI was completely dissolved.

3.3.2 Sample preparation

Potatoes (cv. 'Russet Burbank') purchased from a local supermarket (Supermarchés Provigo, Montreal, QC, Canada) were held at room temperature (~21°C). Russet Burbank potatoes were chosen due to its availability at the time of this study. The Tubers were manually peeled and immediately cut into slices 2.5 - 3.5 mm in thickness. Samples potato strips were weighed, then dipped in the coating suspensions for 20 - 40 seconds, weighted again, and then fried.

Coated and non-coated potato strips were fried using a deep-fat fryer (Delonghi Digital Deep Fryer Model: D24527DZ), filled with 1.5 L of canola oil. Oil was pre-heated and maintained at 180±1°C for all samples. Samples were fried for 60, 120, 180, or 240 s, and then drained by vigorously shaking the fryer basket. To avoid degradation of the oil during frying, each batch of oil was used for 2 hours before it was replaced with a fresh batch of oil. All experiments were performed in triplicate.

3.3.3 Water content

To measure potato slice water content, fried samples were cooled to room temperature, weighed and then dried to constant weight at 105°C in a forced air convection oven (Isotemp 700, Fisher Scientific, Pittsburgh, PA, USA), and the final weight noted. For both coated and uncoated samples, moisture content (MC) was calculated by the gravimetric method (AACC, 1986). Moisture ratio (MR) was also determined as the moisture content at a given time divided by initial moisture content.

3.3.4Fat content

The total fat content *(FC)* of fried samples was determined from dried and finely ground (coffee grinder) material using the standard AOAC procedure (Soxhlet method). A Soxhlet extractor (SER 148, Velp Scientifica, Usmate, Italy) was used to determine fat content in samples. Fat was extracted using petroleum ether as solvent. The fat content gained after frying (*FC*) is expressed on a dry weight basis (i.e., mass of fat extracted from a sample of dry, ground, previously-fried potato divided by the sample's mass).

Film pick up rates were calculated as the difference in weight of NC samples and weight resultant after the adherence of the coating material.

3.3.5 Color measurements

Color intensity of WPI and non-coated samples was obtained using a Konica Minolta colorimeter (Model No: CR-300, Konica Minolta, Sensing, Inc. Osaka, Japan), calibrated with a standard white ceramic plate. Ten measurements were performed on the surface of five coated strips, one measurement on each side of each potato strip, for each formulation batch. Results were expressed in the colorspace of *L*, *a**, *b** chromaticity coordinates (Commission Internationale de l'Éclairage), where *L* is lightness, *a** ranges from red to green, and *b** ranges from yellow to blue (McLaren, 1976). The total color change (ΔE) of each batch was calculated as

$$\Delta E = \sqrt{(L_0 - L)^2 + (a_0 - a)^2 + (b_0 - b)^2} \quad \textbf{(3.1)}$$

where,

L_0, a_0, b_0 are the *L*, *a*, and *b* values for the non-coated potatoes at initial time.

L, a, b are the *L*, *a*, and *b* values for the coated potatoes at a given frying time.

3.3.6 Textural Analysis

The texture of coated potato strips was obtained through compression tests undertaken on an Instron Universal Testing Machine (Model 4502, Canton, MA, USA) using a cylindrical 5 mm diameter probe at 50 N load at a cross head speed of 60 mm min^{-1}. Maximum force (N) and slope of the linear section of load displacement were recorded as hardness and Young's modulus, respectively.

3.3.7 Statistical Analysis

Data were subjected to analysis of variance using SAS System software (Version 9.2, SAS Institute, Inc, 1999, Cary, NC, USA). A General Linear Model (PROC GLM) procedure was conducted for analysis of variance (ANOVA) to determine the significant differences of coated potato strips. Differences among treatments were determined using Duncan's Multiple Range Test at 5% confidence test (or $\alpha = 0.05$). ANOVA results for parameters tested are shown in Appendix 3.1-3.2. Correlation was obtained to relate moisture loss and fat content. Normality of residuals were tested in all data recorded using a PROC UNIVARIATE model. Shapiro-Wilks *W*, Kolmogorov-Smimov *D*, Cramer-vonMises W^2, and Anderson-Darling A^2 were used to assess whether the residuals' distribution deviated

significantly ($\alpha = 0.05$) from normality. Values recorded for tests of normality are displayed in Appendix 3.3. Parameters MC, *a-value* and F_{max} required transformation to establish normality; these transformations were *ln*(MC), *exp*(*a*+2), and *ln*(F_{max}).

3.4 Results and Discussion

3.4.1 Effect of coating formulations on moisture content of fried potato strips.

Film pickup was neither affected by plasticizer type nor by plasticizer concentration ($P > 0.05$). This could by explained due to similarities in viscosity and deposition rate within coating formulations. Higher viscosity is due to higher solids content present in the film, which are influenced by the concentration of film-forming agent and not by the plasticizer applied.

Both coating type [(none), sorbitol (3) and xylitol (3) coatings] and frying time had significant ($P \leq 0.05$) effects on MC. Although significant differences on moisture content ($P \leq 0.05$) were recorded between non-coated (40.69%) and coated samples, no differences were detected within coating formulations (values ranged from 46.07 to 49.02%; Table 3.1).

Consistent with this, the lowest moisture content value was observed in non-coated samples (8.45%) at last minute of frying (240 s). Both coated and non-coated samples followed typical moisture content reduction curve over frying time; however, coated samples tracked lower rates of moisture loss meaning that the formulations were able to retain more water (reduce water evaporation) mostly at first minutes of frying (Figures 3.1-3.2). These data are comparable and support by previous studies, where the incorporation of WPI resulted in relatively large increase of water content on coated samples, therefore greater final moisture content on fried coated samples (Dogan et al., 2005; Dragich & Krochta, 2009).

Table 3-1 Mean values for oil content, moisture content, colors and texture of different WPI coating formulations and non-coated samples after deep fat-frying.

Parameter	Coating Type						
		10% w/w Whey protein (WPI)					
	None	Sorbitol (%)			Xylitol (%)		
		0.5	0.75	1	0.5	0.75	1
MC	40.69b	45.11ab	46.07a	46.91a	48.65a	49.02a	48.72a
FC	20.07a	17.14b	16.54b	17.14b	17.73b	17.97b	17.07b
L	52.67b	54.63ab	53.57ab	54.92a	54.79ab	53.63ab	55.43a
a	4.07ab	3.02ab	2.46b	4.49a	4.42ab	4.00ab	2.55b
b	12.86a	13.19a	13.77a	16.33a	14.44a	13.55a	13.79a
F_{max}	1.44b	1.92b	2.98a	1.75b	1.86b	1.75b	1.87b
λ	0.75b	1.10ab	1.15ab	1.75a	1.30ab	0.94b	0.73b

Mean values are significantly different row-wise within treatment factor if the letters differ (Duncan's multiple range test, α=0.05). L= lightness; a= redness; b= yellowness; F= maximum force; λ= Young's modulus. Mean values throughout the entire frying process.

Moisture ratio (MR) exhibited similar reduction in water rates in all coated samples across frying time ending with higher values than control samples. This could be attributed to the hydrophobic nature the protein association through hydrogen bonding. The potential formation of intermolecular sulphide cross-links in whey protein films could be the responsible for exhibiting improved barrier properties for water vapour (Dogan et al., 2005).

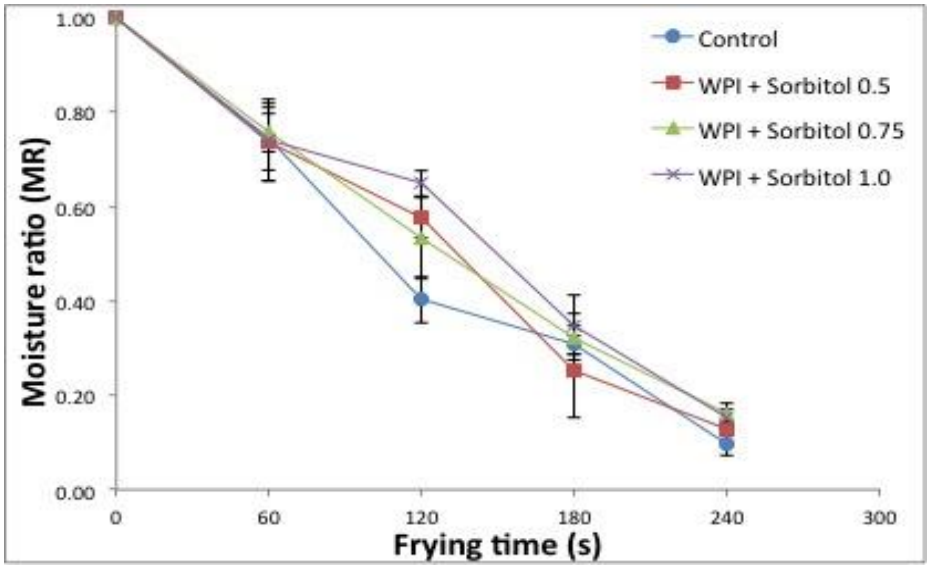

Figure 3-1. Moisture ratio (MR) in fried coated potato strips - WPI sorbitol.

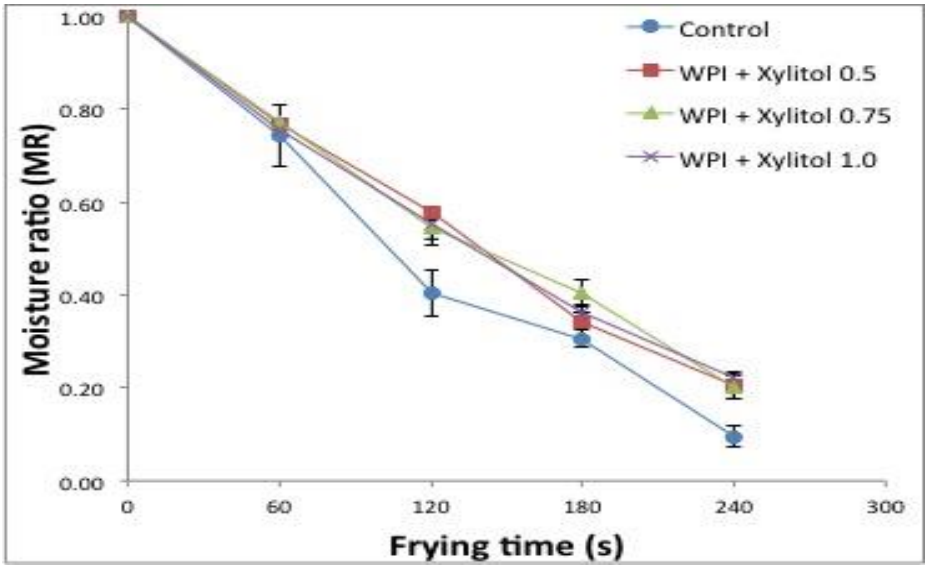

Figure 3-2. Moisture ratio (MR) in fried coated potato strips - WPI xylitol.

Across all coating types, frying duration reduced MC significantly from each frying time to the next, with this difference being of progressively greater extent as frying time went

on. Surprisingly, there was an increase in the moisture loss rate after 120 s of frying, indicating that after this frying time the moisture retention effect of the coating seem to be diminished. WPI-xylitol coating formulations exhibited higher MC values during frying than did sorbitol coatings suggesting that WPI-X formulations to better protect potato strips from moisture loss; however, plasticizer concentration had no effect within a range of 0.5 to 1.0%. In addition, WPI-1.0X coating formulations showed the highest values of moisture ratio (MR = 0.22) at the final frying time (240 s).

3.4.2 Effect of coating formulations on fat content of fried potato strips

Both coating formulations and frying time had significant effects on fat absorption rates [($P \leq 0.05$); (Figures 3.3-3.4)]. Across all frying times, non-coated potato strips gained more fat [(20.07%; Table 3.1)] than did any of the coated strips (16.54-17.97%).

The coatings were significantly effective in reducing fat content by potatoes during frying. Uncoated samples at 240 s showed highest rates of oil content (FC = 27%). Less oil absorption can be related to the formation of covalent links within films during heating, as well as for thermal gelation properties and film-forming ability of proteins (Dogan et al., 2005). In addition, WPI coatings may have modified the surface structure of the product by filling the pores, therefore reduced both trapped oil and surface area through which oil diffused (Gamble et al., 1987; Gamble & Rice 1988; Saguy & Pinthus 1995).

The rates of fat absorption in coated samples were clearly lower than non-coated samples. However, the trends of fat up-take in WPI-sorbitol coating samples over frying time were marginally different. This implies that the different sorbitol concentrations applied (0.5-1.0%) as plasticizers in the coating formulation did not caused a major effect in the rates of fat absorption.

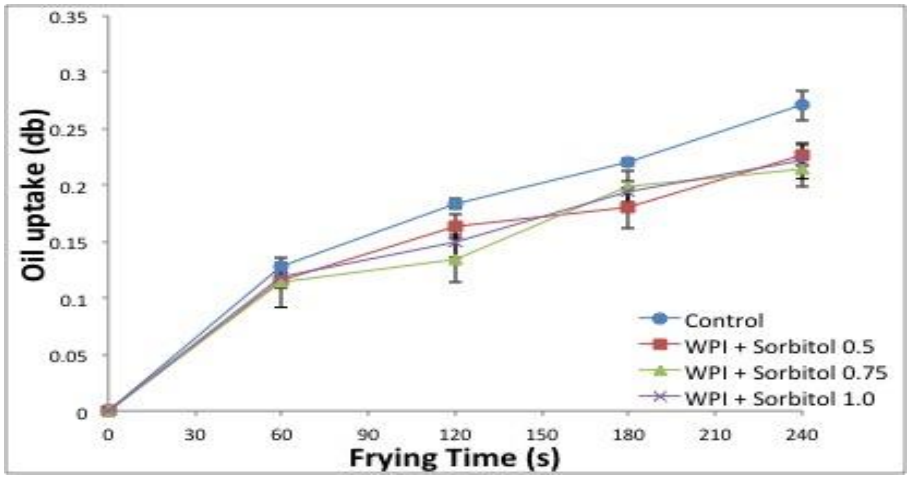

Figure 3-3. Mean oil content in fried coated potato strips - WPI sorbitol.

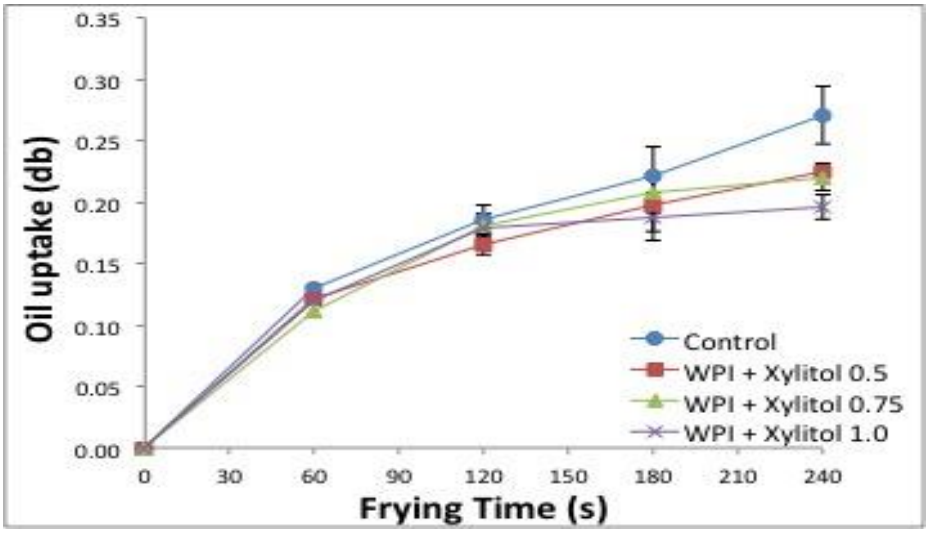

Figure 3-4. Mean oil content in fried coated potato strips – WPI xylitol.

The WPI-1.0X coating formulation resulted in the least fat content (27.72 ± 6.65%) at 240 s frying time (Figure 3.4). This reduction is lower than or comparable to, past reductions in fat absorption using WPI as coatings in fried products. Adequate concentrations in coating formulation, differences in adhesion between coating suspension and substrate applied, proper characteristics of sample, and frying time can be the causes responsible for oil up-take reduction (Garmakhany et al., 2008). Furthermore, WPI-1.0X exhibited to reduce water evaporation exhibiting greatest MC values at 240 s frying time. This could be attributed to the effectiveness of the coating formulation acting a barrier decreasing mass transfer during deep-fat frying.

The oil content was found to be correlated with the moisture content within coated and non-coated samples. This was observed (Table 3.2) in agreement with the work of Gamble et al. (1987) who reported a similar relationship when examining mass transfer in fried potato strips. Mass transfer is controlled by diffusion as a function of frying time and temperature. As moisture migrates outwards, an opposite inflow of oil up-take take place by diffusion to fill up the pores formed by moisture loss (Ngadi et al., 2008).

Table 3-2. Relationship between moisture loss (ML) and fat content (FC) for WPI coated and non-coated potato slices.

Plasticizer		*%ML vs. %FC*	
Type	Concentration (%)	Equation	R^2
No coating		$\%ML = 2.81(\%FC) - 4.61$	0.96
Sorbitol	0.50	$\%ML = 3.22(\%FC) - 5.86$	0.88
	0.75	$\%ML = 3.16(\%FC) - 4.87$	0.94
	1.00	$\%ML = 3.08(\%FC) - 5.91$	0.93
Xylitol	0.50	$\%ML = 2.89(\%FC) - 5.91$	0.92
	0.75	$\%ML = 2.73(\%FC) - 4.54$	0.91
	1.00	$\%ML = 2.91(\%FC) - 4.66$	0.85

3.4.3 Effect of coating formulations on color development of fried potato strips.

Color development in fried potatoes only begins when a sufficient amount of drying has occurred, depending substantially on the drying rate and the heat transfer coefficient during the different stages of frying (Pedreschi & Zuñiga, 2009). Significant changes in color parameters (*L, a, b, ΔE*) occurred as frying proceeded ($P \leq 0.05$). As lower values of *L* represents a darker color one would expect the *L* value to decrease as frying time increased. This was indeed observed (Figure 3.5), with a steady and significant ($P \leq 0.05$) decrease in *L* from each frying time to the next in accordance with previous studies (Krokida et al., 2001; Ngadi et al., 2007).

Coated samples with the highest concentrations of plasticizers: 1.0% xylitol and sorbitol exhibited the highest *L* values (53.47 ± 0.46; and 51.78 ± 2.35, respectively), which were significantly greater than those of non-coated samples, but did not differ significantly from the other coating formulations. In general, the coatings led to greater *L*-values compared to non-coated samples suggesting that the samples were lighter (less darker) than non-coated

potato strips. Improved color quality is associated with high *L*-value (Habib & Brown, 1956; Marquez & Añon, 1986).

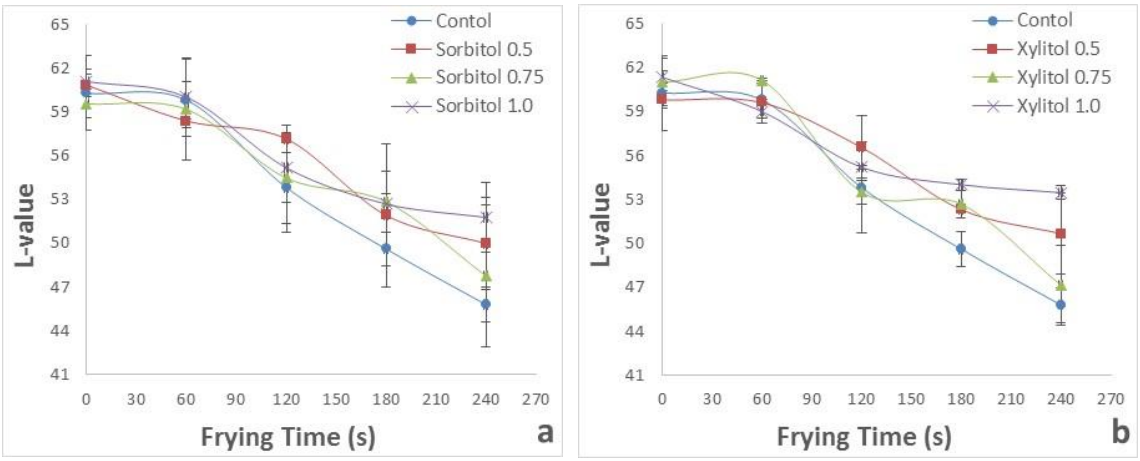

Figure 3.5. L-values in fried coated potato strips - WPI sorbitol (a) and xylitol (b).

The color parameter *a** (red-green) was significantly influenced by frying process time. Across coating types, the *a* value increased significantly from a negative value at 0 s frying, to significantly greater positive values at subsequent times. The *a** values increased significantly from the 120 to the 180 s frying time, but the *a** value for the 240 s frying was intermediate between the two and not significantly different from either (Figure 3.6). Thus in general, as frying progressed the potato slices shifted towards a redder color. Fried potato strips tend to get darker (redder) as frying proceeds' due to non-enzymatic browning reactions, resulting in a progressive increase in *a** values (Pedreschi et al., 2005).

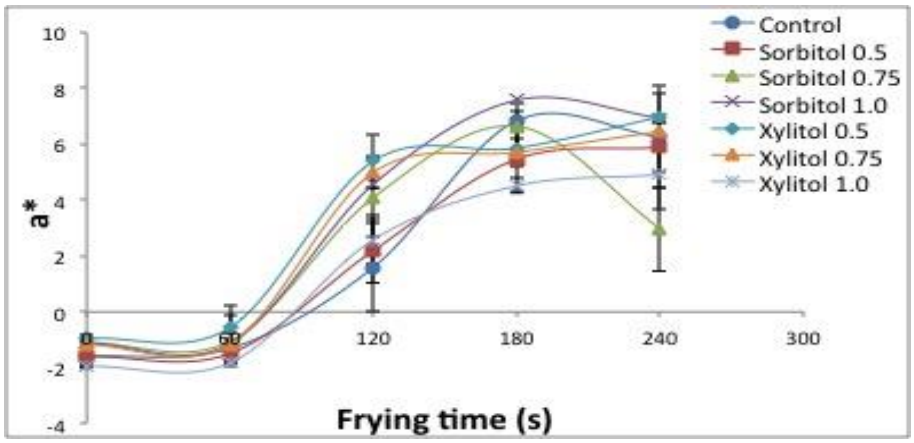

Figure 3.6. a*-values (red-green) in fried coated potato strips for both WPI sorbitol and xylitol formulations**.**

The color parameter *b* (yellow-blue) was not affected by the coating type, but did increase with frying time till reached 180 seconds of frying [($P \leq 0.0001$); (Figure 3.7)], but remaining at similar levels thereafter. Positive values of *b* are desirable in fried potatoes, and increase when temperature and frying time are raised (Krokida et al., 2001). Color development only begins when enough drying has occurred in potato strips — depending upon drying rate — that changes occur in the potato surface (Pedreschi et al., 2005b).

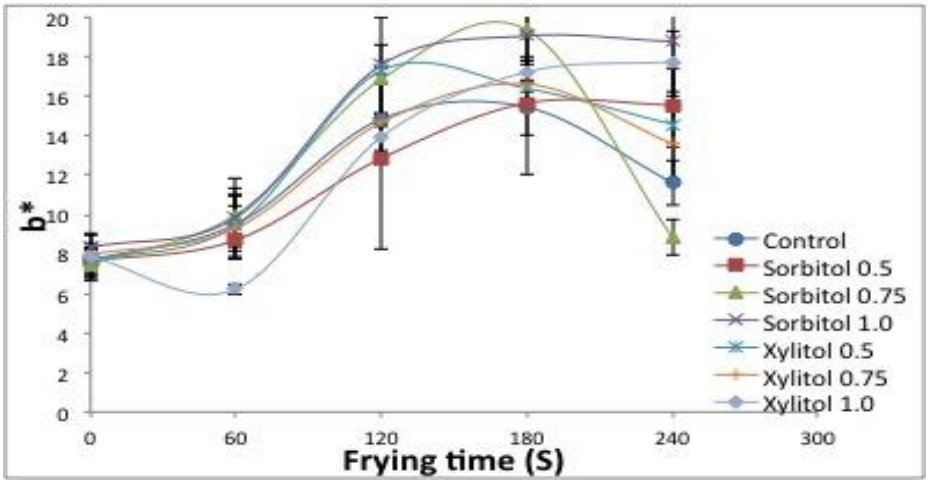

Figure 3.7. b*-values (yellow-blue) in fried coated potato strips for both sorbitol and xylitol formulations.

The color parameter ΔE was significantly ($P \leq 0.001$) affected by frying time but not by type of plasticizer ($P > 0.05$). The parameter reached the highest value at 240 seconds. Frying time had a strong effect on the development of color in all treatments assessed ($P \leq 0.05$), but the presence or absence of coating material or the nature of the coating material itself had relatively little influence on color development in agreement with studies similar results were recorded (Ballard, 2003).

In general, color development was influenced by starch gelatinization, protein denaturation and by non-enzymatic browning of the crust (Sahin, 2000). The presence of lactose from whey protein coatings can be also involved in browning reactions that impart more color to the food material (Kulp & Loewe, 1990).

3.4.4 Effect of coating formulations on mechanical properties of fried potato strips

The force at maximum load (F_{max}) was significantly influenced by frying time ($P \leq 0.0001$). While the non-coated potato strips tended to show lower F_{max} mean throughout the frying process values (1.44 N) than coated strips (1.75-2.98N) this difference was not significant; however, the 0.75% sorbitol formulation showed a significantly greater F_{max} (2.98N) than any other coating ($P \leq 0.05$). The F_{max} value increased significantly from one frying time to the next, up to the entire 240 (s) frying. When F_{max} was expressed relative to non-coated samples it was found to vary significantly with frying time ($P \leq 0.05$), and differed marginally ($P \leq 0.075$) with plasticizer type, but not concentration ($P > 0.30$). The use of sorbitol as plasticizer led to a greater $F_{max,}$ (*i.e*., a greater improvement over the non-coated samples) than the use of xylitol. The relative increase in F_{max} was greatest at 180 (s), and was least at 60 and 240 seconds. In general, the use of a coating improved hardness over non-coated samples, and sorbitol contributed greater hardness than xylitol, though in both cases the difference was statistically marginal. As expected, the F_{max} increased with frying time, a little under 6-fold between 60 and 240 seconds of frying.

Potato strips fried at shorter frying times' exhibited softer external surface due to lamella media solubilisation and starch gelatinization. This can be explained due to the formation of crust on the external surface of fried potatoes (Pinthus et al., 1995). The gradual increase of potato strips' F_{max} over frying time was the result of a reduction in moisture content and the development of an external crust (Figure 3.8). Similar results were recorded in previous studies in fried potato strips (Garmakhany et al., 2008).

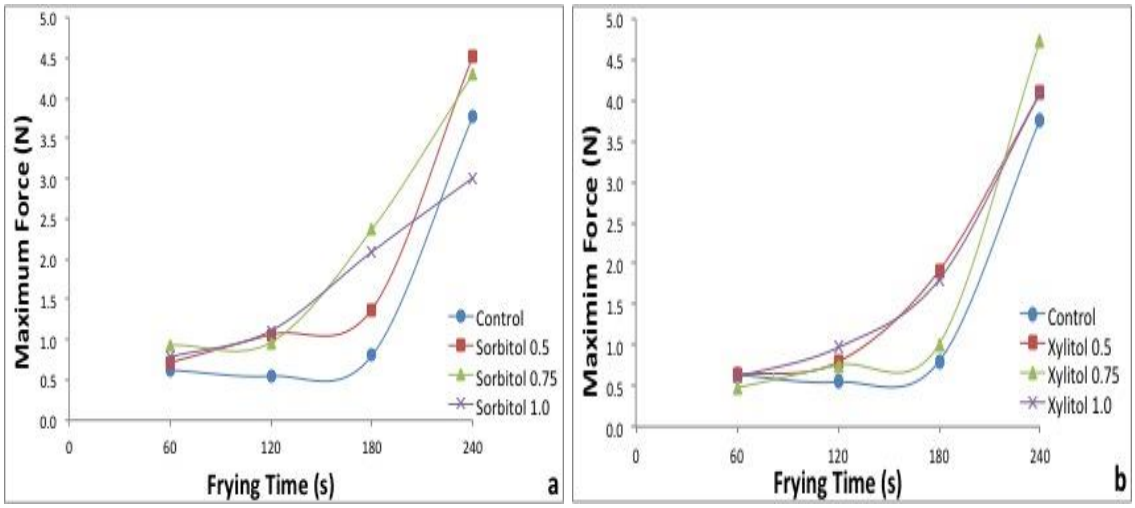

Figure 3.8. F_{max} (N) values in fried coated potato strips - WPI sorbitol (a) and xylitol (b).

Young's modulus (λ), as a function of the stiffness of fried samples was also determined. Marginally significant differences ($P \leq 0.075$) were noted for the type of plasticizer and concentrations (none + 6 coatings) but the differences (Duncan's, $P \leq 0.05$) showed no clear pattern amongst them between coated and uncoated samples ($P \leq 0.05$).

The value of λ increased significantly from the first three frying times (0.41-0.76 N mm^{-1}) to the 240 seconds frying time (2.79 N mm^{-1}). As with the F_{max} the mechanical properties changed substantially over the last minute of frying (180 *vs.* 240 s), indicating that much of the desirable mechanical properties were developed at the end of the frying process. The value of λ was significantly higher at 180 seconds than at any other frying times, and these did not differ significantly amongst themselves.

3.5 Conclusions

Whey protein coating formulations demonstrated exceptional mass transfer barrier properties. Coating formulations with xylitol added as a plasticizer showed greater rates of MC after frying. Therefore, they act as better barriers for the reduction of moisture loss rates during frying primarily at 60-120 seconds of frying. Non-coated samples gained more total fat content (%*FC*) than did any of the coated potato strips. Moreover, the nature of the plasticizer applied demonstrated to have significant influence in total fat content. WPI-1.0X exhibited the most suitable mass transfer barrier properties, exhibiting greatest both moisture content *(%MC),* and total reduction of oil content (18.46; 27.72 %), at last minute of frying (240 s), respectively. The oil up-take was found to be correlated with the moisture loss within coated and non-coated samples exhibiting higher coefficients of correlations ($R^2 \geq 0.85$).

Color development demonstrated to begin as frying preceded decreasing L-values as frying continued. The presence or absence of coating materials in fried potato strips demonstrated to have relatively low influence on color development with the exception of (1.0%) plasticizer concentration, which samples exhibited to be lighter than control. Mechanical properties demonstrated to increase over frying time for both coated and non-coated samples. Coated potato strips tend to exhibit greater F_{max} values than non-coated strips; however, these differences were not significant. The gradual increase of potato strips' F_{max} as a function of hardness over frying time was the result of a reduction in moisture content and the development of an external crust. Young's modulus as a function of stiffness in coated potato strips showed no significant differences with respect to non-coated samples. The value of λ increased significantly over frying time. Moreover, frying had significant effects on all variables analyzed (%*MC* %*FC,* L, a, b, ΔE, $F_{max,}$ and λ).

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CONNECTING TEXT

In chapter III, the influence of whey protein isolated coatings plasticized with sorbitol and xylitol on moisture loss, fat absorption, color development and mechanical were recorded. The effect of frying time was significant to all the variables studied among coated and non-coated potato strips. Whey protein isolated coating formulations influenced positively on the overall quality of coated potato strips during deep-fat frying. Mass transfer phenomena noted reduced rates, improved color development and mechanical properties were also observed in all fried samples tested. Polysaccharide based coatings has been extensively studied during the last few years. In particular, it was noted that methylcellulose coatings are suggested to exhibit the best barrier properties for fried products. It is therefore necessary to study both (whey protein isolated and methylcellulose) coatings to determine their functionality as coatings to improve quality in coated potato strips during deep-fat frying. In chapter IV, methylcellulose based coatings were studied following the methodology applied in chapter III. Effects on moisture loss, fat absorption, color development and mechanical properties were also evaluated.

IV. INFLUENCE OF METHYLCELLULOSE-BASED COATINGS ON QUALITY OF POTATO STRIPS DURING DEEP-FAT FRYING.

4.1 Abstract

Over the past years growing demands for low-fat products, especially for those low in saturated fats, have been growing in order to reduce the risk of coronary. Thus, reducing fat content of fried products has become a priority. The application of coating suspensions is one effective way to meet consumers' preferences and health concerns. A methylcellulose-based coating composed of 1% methylcellulose (MC), plasticized with sorbitol (S) or xylitol (X) at concentrations of 0.50, 0.75 or 1.00, were applied to potato strips prior to frying. Oil and moisture transfer, color development and changes in textural properties after 1, 2, 3, or 4 min of frying were assessed in non-coated (control) and coated (MC.) fried potato strips. Moisture loss (*ML)* during frying was significantly ($P \leq 0.05$) increased in non-coated samples. Although the type of the plasticizer did not show any significant difference, the concentration each plasticizer did have a significant effect on *ML*. Formulation with methylcellulose and 0.50% Sorbitol (MC-0.50S) showed greatest values of moisture content (17.83%) at the last minute of frying (240 s). As frying proceeded non-coated samples gained significantly ($P \leq 0.05$) greater fat content values than any of the coated slices. Total fat content *(FC)* in non-coated samples at 240 s of frying showed the highest rates of oil absorption (36.99%), while in coated potato strips *(FC)* significantly decreased compared to non-coated samples and each coating formulation resulted in different rates of oil content. The most suitable methylcellulose coating formulation in terms of reduction of oil uptake was MC-0.5S at 240 s frying time. Regarding color development *L*-values was significantly ($P \leq 0.05$) higher for coated than for non-coated samples as frying proceeded. Mechanical properties increased over frying time for both coted and non-coated samples. Moreover, load at maximum load (F_{max}) as a function of hardness and Young's modulus (λ) as a function of stiffness recorded a significant increased after 180 s of frying, indicating that most of the desirable textural properties were developed at the end of the frying process once enough water evaporation and crust formation has occurred in both coated and non-coated potato strips. Frying time

had significant effects ($P \leq 0.05$) on all variables relative to non-coated samples analyzed (MR, *%FC*, *L*, *a*, *b*, $F_{max,}$ and λ).

4.2 Introduction

Deep-fat frying is a widely used method for preparing foods with an attractive and tasty surface. The soft and moist interior along with porous crispy crust improves palatability to foods (Mallikarjunan et al., 1997). Furthermore, fried foods exhibit attractive aromas and visual appeal due to the golden-brown color. Development of color during frying is a major factor of consumer acceptance (Kulp & Loewe, 1990). Potato (Solanum tuberosum) is one of the world major agricultural crops consumed by millions of people worldwide. Moreover, potatoes are grown in approximately 80% of all countries in the world (Durán et al., 2007). Production of desirable characteristics in fried products are influenced on heat capacity of the frying medium, thermal conductivity of the food item, appropriate temperature differential between the oil medium and food, dehydration of the surface, and oil interaction with food components to obtain the desired texture and flavour (Mellema, 2003).

Deep-fat frying is a complex process linking simultaneous heat and mass transfer (Suarez et al., 2008). It consists in the total immersion of food item into hot oil; the high temperature causes evaporation of water, which is released from the food item through the surrounding oil. Oil is absorbed by the food, replacing some of the lost water (Mellema, 2003). When products with relatively high starch content (i.e. potatoes) are heated; their textural properties suffer changes caused mainly by gelatinization of the starch content (Pedreschi & Moyano, 2005). Textural attributes of fried potatoes include firmness, hardness, and elastic modulus; where firmness has been related to the sense of chewing (Nourian et al., 2003). Changes in texture of fried potatoes are influenced by changes in the cellular and sub cellular structure of the products (Odenigbo et al., 2012).

Stiffness depends on mechanical properties of the material and structural elements (e.g., air cells, micro-heterogeneities, geometrical parameters, etc.) The maximum force recorded before a rupture in the structure is related to hardness of the material (Miranda & Aguilera, 2006). Finished fried potato strips exhibit a structure made of hard crust region and a soft-core region (Pedreschi et al., 2001).

Fried potato color is the result of Maillard reaction that depends upon the formation of brown pigments of the potato slices in oil, caused by a reaction between sugars and amino acids (Marquez & Añon, 1986). The appearance and color of the food surface is the first quality parameter evaluated by consumers being critical in the acceptance of the final product (Pedreschi et al., 2005b).

Despite the shift in consumer preferences to low-fat foods, people still consume high levels of fried products, which are linked to coronary diseases and obesity. Thus, reducing fat content of fried foods by the application of coatings has become an alternative to meet consumer preferences and health concerns (Garcia et al., 2004). Several authors have studied the properties of different coatings to reduce oil absorption (Mackinson et al., 1987; Debeaufort & Voilley, 1997; Mallikarjunan et al., 1997; Williams & Mittal, 1999; Garcia et al., 2004).

Coatings regulate the transfer of moisture, oxygen, carbon dioxide, lipid, aroma, and flavour compounds in the food systems (McHugh et al., 1994). Moreover, coatings enhance the quality of food products protecting them from physical, chemical and biological deterioration, acting as barriers against oils, gases, or vapours (Han & Gennadios, 2005; Kester & Fennema, 1986).

Coatings can exhibit hydrophilic or hydrophobic properties; they are composed of natural film-forming materials such as hydrocolloids, proteins, lipids and combinations of these three. Polysaccharides materials especially cellulose derivate such as methylcellulose (MC), carboxy methylcellulose (CMC), hydroxypropyl cellulose (HPC) and hydroxypropyl methylcellulose (HPMC) are the most common polysaccharides materials used for coating formation.

According to Li et al. (2002), cellulose based coatings can form strong intermolecular hydrogen bonds; however, original cellulose is insoluble in water. The replacement of hydroxyl groups for hydrophobic groups such as methyl or hydroxypropyl groups, to create water-soluble cellulose compounds it is necessary. Methylcellulose thermal gelation can take place in the temperature range about 50 to 70 °C due to hydrophobic association. Contrarily, hydrophobically modified cellulose is thermoreversible in hydrophobic association, which means that the associate aggregates formed at elevated temperatures can return to liquid state again upon cooling through the

disassociation of hydrophobic groups (Li et al., 2002). Maskat et al. (2005) the thermal gelling ability of MC and HPMC provide structural integrity to a batter before and after frying; thermal gelation encourages cohesion and adherence to the surface of the substrate.

MC films forms gels, where hydrophobic polymer chain interactions are involved in the thermal gelation process (Zaritzky, 2011). Thermal gelling properties in MC coatings encourage cohesion and adherence (film pick-up) to the surface of the substrate, where viscosity of coating formulations as a rheological property has an important role in the proper adherence of the material (Tavera-Quiroz et al., 2012).

Williams and Mittal (1999) found that MC coatings were best barrier properties in terms of reduction of oil up-take in several food products tested (frozen potatoes, rehydrated potato starch, mashed potatoes and commercial pastry mixes). Moreover, Mallikarjunan et al. (1997) proved that mashed potato balls coated with MC exhibit 83.6% of reduction on fat absorption determining to be the best suitable material for coating formations. This work focused on studying the efficacy of sorbitol and xylitol as plasticizers of methylcellulose coatings used to record quality attributes (moisture loss, fat content, color development and mechanical properties) during deep-fat frying of coated potato strips.

4.3 Materials and Methods

4.3.1 Hydrocolloid solutions

Coating ingredients included methylcellulose M352-500 (Fisher Scientific S.A New Jersey, USA), sorbitol (BP439-500; (Fisher Scientific, ON, Canada) and xylitol (99% AC22598-0250; Fisher Scientific, ON, Canada). Aqueous suspensions of 1% MC were used. Methylcellulose suspensions were dissolved in hot water (100 °C). High temperatures promote hydrophobic polymer chain interactions that are involved in the thermal gelation process, especially intermolecular hydrogen bonding within the cellulose ether (Zaritzky, 2011). Sorbitol and xylitol plasticizer concentrations of 0.50, 0.75, and 1.00% (w/w) were tested, resulting in six coating formulations: MC-sorbitol 0.5% (MC-0.5S), MC-sorbitol 0.75% (MC-0.75S), MC-sorbitol 1.0% (MC-1.0S), MC-xylitol 0.5% (MC-0.5X), MC-xylitol 0.75% (MC-0.75X), MC-xylitol 1.0% (MC-1.0X). The MC (1g) was weighed and slowly dispersed in 30 mL of hot distilled water at 100 °C under constant

stirring. Once a homogeneous solution was obtained (approximately 1 h), total volume was completed to 100 mL with cold distilled water was added to the solution under stirring until room temperature was reached. Plasticizers were incorporated after MC was completely dissolved.

4.3.2 Sample preparation

Potatoes (cv. 'Russet Burbank') purchased from a local supermarket (Supermarchés Provigo, Montreal, QC, Canada) were held at room temperature (~21°C). Russet Burbank potatoes were chosen due to the availability of it during the time the research was conducted. Tubers were manually peeled and immediately cut into slices 2.5 - 3.5 mm in thickness. Samples potato strips were weighed, then dipped in the coating suspensions for 20 - 40 s, weighted again, and then fried.

Coated (MC) and non-coated control (NC) groups were fried using a deep-fat fryer (Delonghi Digital Deep Fryer Model: D24527DZ), filled with 1.5 L of canola oil. Oil was pre-heated and maintained at 180±1°C for all samples. Samples were fried for 60, 120, 180, or 240 s, and then drained by vigorously shaking the fryer basket. To avoid degradation of the oil during frying, each batch of oil was used for 2 hours before it was replaced with a fresh batch of oil. All experiments were performed in triplicate.

4.3.3 Water content

To measure potato slice water content, fried samples were cooled to room temperature, weighed and then dried to constant weight at 105°C in a forced air convection oven (Isotemp 700, Fisher Scientific, Pittsburgh, PA, USA), and the final weight noted. For both coated and uncoated samples, moisture content (MC) was calculated by the gravimetric method (AACC, 1986). Moisture ratio (MR) was also determined as the moisture content at a given time divided by initial moisture content.

4.3.4 Fat content

The total fat content *(FC)* of fried samples was determined from dried and finely ground (coffee grinder) material using the standard AOAC procedure (Soxhlet method). A Soxhlet extractor (SER 148, Velp Scientifica, Usmate, Italy) was used to determine fat

content in samples. Fat was extracted using petroleum ether as solvent. Oil content was measured on a dry weight basis (db) by dividing the mass of oil extracted from the ground sample by the mass of the dried sample. The fat content gained after frying *(FC)* is expressed on a dry weight basis (i.e., mass of fat extracted from a sample of dry, ground, previously-fried potato divided by the sample's mass).

Film pick up rates were calculated as the difference in weight of NC samples and weight resultant after the adherence of the coating material.

4.3.5 Color measurement

Color intensity of Mc and NC samples was obtained using a Konica Minolta colorimeter (Model No: CR-300, Konica Minolta, Sensing, Inc. Osaka, Japan), calibrated with a standard white ceramic plate. Ten measurements were performed on the surface of five coated strips, one measurement on each side of each potato strip, for each formulation batch. Results were expressed in the colorspace of *L*, *a**, *b** chromaticity coordinates (Commission Internationale de l'Éclairage), where L is lightness, *a** ranges from red to green, and *b** ranges from yellow to blue (McLaren, 1976). The total color change (ΔE) of each batch was calculated as

$$\Delta \boldsymbol{E} = \sqrt{(\boldsymbol{L_0} - \boldsymbol{L})^2 + (\boldsymbol{a_0} - \boldsymbol{a})^2 + (\boldsymbol{b_0} - \boldsymbol{b})^2} \qquad \textbf{(4.1)}$$

where,

L_0, a_0, b_0 are the *L*, *a*, and *b* values for the non-coated potatoes at initial time.

L, a, b are the *L*, *a*, and *b* values for the coated potatoes at a given frying time.

4.3.6 Texture Analysis

The texture of coated potato strips was obtained through compression test undertaken on an Instron Universal Testing Machine (Model 4502, Canton, MA, USA) using a cylindrical 5 mm diameter probe at 50 N load and cross head speed of 60 mm min^{-1}. Maximum force (N) and slope of the linear section of load displacement were recorded as hardness and Young's modulus, respectively.

4.3.7 Statistical Analysis

Data were subjected to analysis of variance using SAS System software (Version 9.2, SAS Institute, Inc, 1999, Cary, NC, USA). A General Linear Model (PROC GLM) procedure was conducted for analysis of variance (ANOVA) to determine the significant differences of coated potato strips. Differences among treatments were determined using Duncan's Multiple Range Test at 5% confidence test (or $\alpha = 0.05$). ANOVA results for parameters tested are shown in Appendix 4.1-4.2. Correlation was obtained to relate moisture loss to fat content.

Normality of residuals were tested in all data recorded using a PROC UNIVARIATE model. Shapiro-Wilks *W,* Kolmogorov-Smimov *D,* Cramer-vonMises $W^{2,}$ and Anderson-Darling A^2 were used to assess whether the residuals' distribution deviated significantly ($\alpha = 0.05$) from normality. Values recorded for tests of normality are displayed in Appendix 4.3.

4.4 Results and Discussion

4.4.1 Effect of coating formulation on moisture content of fried potato strips.

Film pick-up rates were not affected by the type of plasticizer applied nor on the concentration added in the coating formulation ($P > 0.05$). Initial moisture content in non-fried coated samples exhibited a slight variation across coating formulations, but it was consistently about 0.45% greater than that of non-coated samples, this difference (83.30% *vs.* 82.85%) was not statistically significant ($P > 0.05$). The formulation MC-0.5S exhibited the highest initial moisture content rates (83.31 ± 0.17%).

Both type of plasticizer applied in the coating formulation [(none), (3) sorbitol and (3) xylitol coatings)] and frying time had significant ($P \leq 0.05$) effects on MC. Mean MC values through the entire frying procedure showed that non-coated potato strips were marginally less (37.22%) than that of coated samples (Table 4.1).

Consistent with this, MC values were significantly lower for non-coated (6.90%) than coated potato treatments (12.57-17.83%) at the 240 s frying time. This can be explained as due to the significant effect of the plasticizer concentration applied. The lowest concentration of the plasticizer yield the highest MC values showing improved moisture transfer barrier and retaining more water (decreasing mass transfer) mostly at the

early frying stage. The rate of moisture loss was observed to decrease significantly as frying proceeded in agreement with the work of Moreira et al. (1999). As water retention is strongly affected by some additives, incorporating cellulose could play a major role in changing the amount of moisture loss and oil uptake (Saguy & Pinthus, 1995).

Table 4-1 Mean values for oil uptake, moisture content, color and texture of different MC coating formulations and non-coated samples after deep fat-frying.

Parameter	Coating Type						
		1% w/w Methylcellulose (Mc)					
	None	Sorbitol (%)			Xylitol (%)		
		0.5	0.75	1	0.5	0.75	1
MC	37.22c	42.89a	41.64ab	41.46ab	41.08ab	39.07bc	38.54bc
%*FC*	28.18a	18.193e	20.372d	22.024cd	21.319cd	22.877c	25.05b
L	48.75bc	53.05a	54.30a	52.06ab	51.61b	51.39b	51.18b
a	3.675a	3.367a	3.672a	4.615a	3.505a	3.570a	3.000a
b	11.01a	12.11ab	12.568a	13.018a	10.94ab	10.66ab	9.77ab
F_{max}	0.88ab	0.722b	0.712b	0.727b	1.22ab	1.572ab	1.097ab
λ	1.355b	1.485b	1.532b	1.607b	1.715ab	2.110a	1.360b

Mean values are significantly different row-wise within treatment factor if the letters differ (Duncan's multiple range test, α=0.05). L= lightness; a= redness; b= yellowness; F= maximum force; λ elastic modulus. Mean values within the entire length of frying procedure.

Non-coated samples had the lowest values of moisture ratio (MR) than in any coated potato strips across all frying times recorded, these differences were more evident at the 240 s frying time (Figures 4.1- 4.2). Coated samples ranged (0.21 to 0.15%) values vs. 0.08 recorded for control samples. The concentration of the plasticizer applied had significant effects on the MR values.

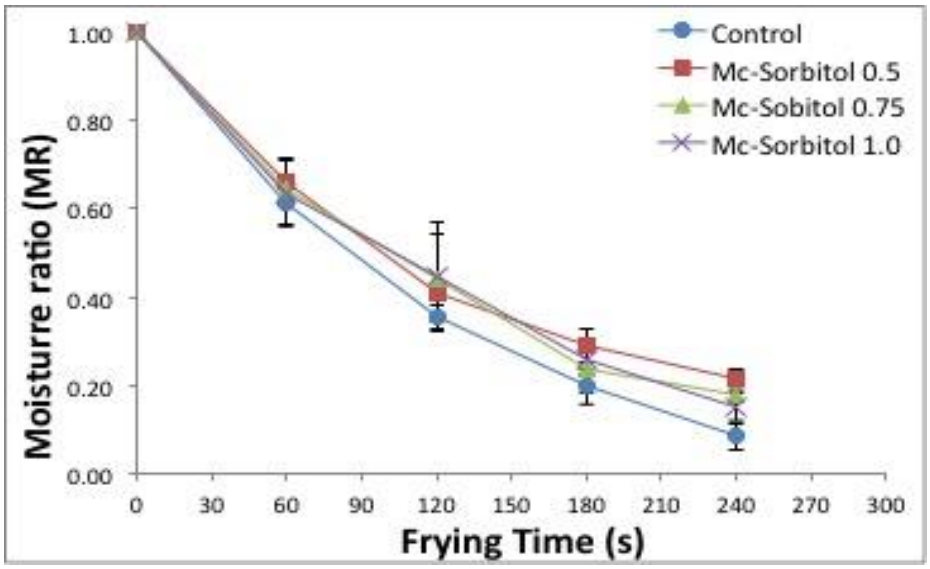

Figure 4-1. Moisture ratio (MR) in fried coated potato strips – MC-S.

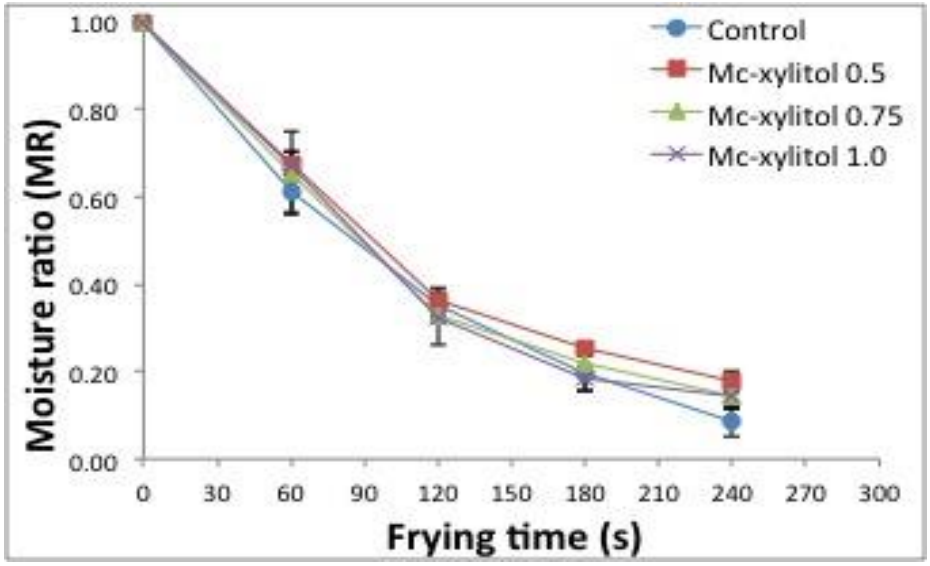

Figure 4-2. Moisture ratio (MR) in fried coated potato strips – MC-X.

Across all coating formulations, the frying time reduced MR significantly from each frying time to the next recording lowest values at the 240 s frying time. Conversely, moisture loss declined significantly from one frying time to the next, with a lessening extent as frying time continued. Greatest moisture loss rates were recorded at 120 s of frying, and least at later times, indicating that the barrier on moisture transfer of the coating material diminished after 120 s of frying. Similarly, Gamble et al. (1987) stated that the

rate of drying decreased at below 20% moisture (180 s frying time) and slowed to the final moisture content.

The potato strips coated with MC-0.5S recorded highest rates of MR at the final frying time [(240 s); (0.21%)]. The differences recorded can be attributed to chemical similarity between coating suspensions on the surface of the sample play a crucial role limiting mass transfer during frying. In addition to a positive adhesion of the coating material in the surface of samples, which is often linked to the appropriate viscosity displayed by the coating formulation (Huse et al., 1998).

4.4.2 Effect of coating formulations on fat content of fried potato strips.

Both coating and frying time had significant effects on fat content ($P \leq 0.05$). Across all frying times, non-coated potato strips gained more fat content (FC = 28.18%) than any of the coated strips [(18.19-25.05%); (Table 4.1)]. The coating formulation had significant effect on total fat content ($P \leq 0.05$). Thus, the coatings were effective in reducing fat absorption by coated potatoes during frying. Oil absorption increased progressively as frying time went on (Figures 4.3-4.4). Uncoated samples at 240 s of frying showed highest rated of oil content (FC = 33%). The most suitable methylcellulose coating formulation regarding the reduction of fat content in percentage was MC-0.5S (FC = 36.99%) at 240 s frying time. These results agreed with those reported by Garcia et al. (2004) who recorded values of 35.2% in potato strips coated with methylcellulose and sorbitol at 0.5%. The efficacy of plasticized MC coatings can be attributed to the thermo-gelling properties exhibited by cellulose materials that could lead to stronger coatings. Moreover, thermo-gelling promote the formation of small amounts of wide punctures with low capillary pressures (Garmakhany et al., 2008).

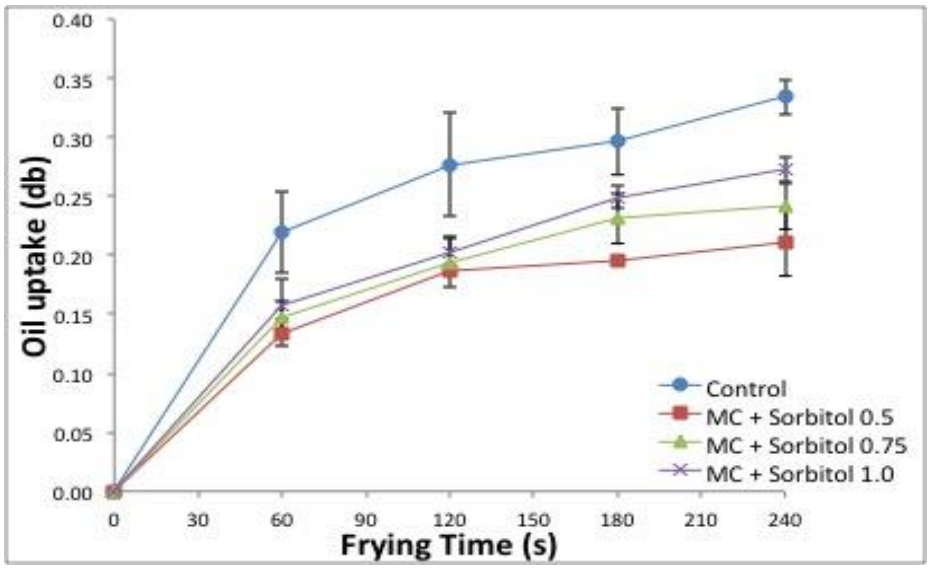

Figure 4-3. Mean oil content in fried coated potato strips MC-S.

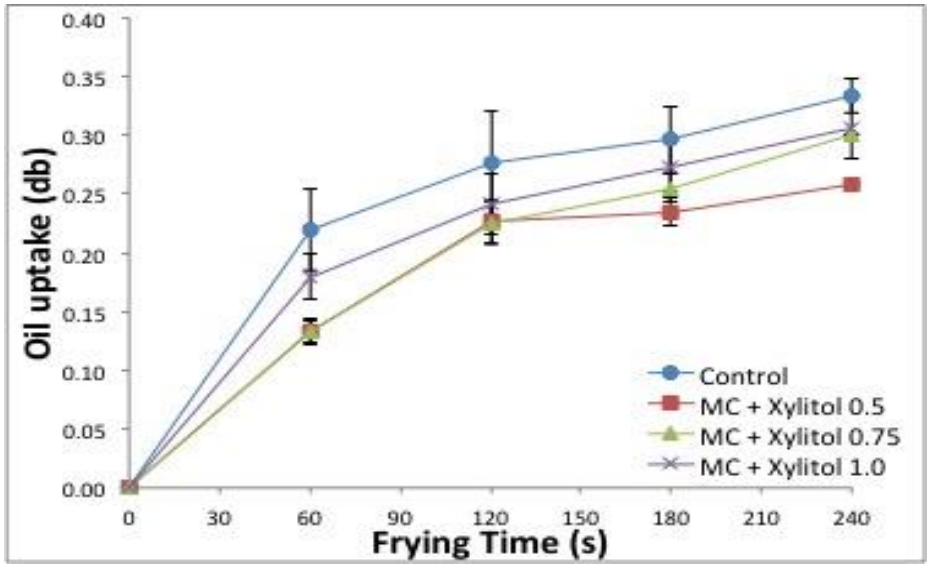

Figure 4-4. Mean oil content in fried coated potato strips MC-X.

Dehydration in hot oil at temperatures from 160 to 180 °C is characterized by high drying rates, which is also related for ensuring favourable textural properties of the final product. However, dehydration leads to substantial increased in fat content (Baumann & Escher, 1995). Heat is transferred from the oil to the food, water is evaporated from the food and oil is absorbed in it (Krokida et al., 2000). Mass transfer has been characterized

by the movement of oil into the product and water, in the form of vapour, form the product (Farkas et al., 2000). In this study, a linear relationship of oil content and moisture loss for the different coating and in the absence of a coating material was recorded (Table 4.2) in agreement with previous reported work (Gamble and Rice 1987; Moreira et., 1997; Krokida et al., 2000).

Pinthus and Saguy (1993) demonstrated that interfacial tension significantly affected oil uptake in deep fat frying of potato strips, suggesting that the mechanism for oil absorption is due to capillary forces. McDonough et al. (1993) concluded that the oil diffused into tortilla chips through small channels formed as water evaporated from the product. As moisture migrates outwards, an opposite inflow of oil up-take take place by diffusion to fill up the pores formed by moisture loss (Ngadi et al., 2008).

Table 4-2. Relationship between moisture loss (ML) and fat content (FC) for MC coated and non-coated potato slices.

Plasticizer		*%ML vs. %FC*	
Type	Concentration (%)	Equation	R^2
No coating		$\%ML = 2.21(\%FC) - 4.20$	0.93
Sorbitol	0.5	$\%ML = 3.01(\%FC) - 3.36$	0.95
	0.75	$\%ML = 2.77(\%FC) - 3.68$	0.96
	1.00	$\%ML = 2.55(\%FC) - 3.26$	0.97
Xylitol	0.5	$\%ML = 2.63(\%FC) - 2.71$	0.98
	0.75	$\%ML = 2.47(\%FC) - 1.01$	0.99
	1.00	$\%ML = 2.43(\%FC) - 4.09$	0.94

4.4.3 Effect of coating formulations on fat content of fried potato strips

The development of color parameters (*L, a, b)* was observed as frying proceeded in potato strips. *L*-values is the luminance or lightness component, ranges from 0 to 100, and parameters a* (green to red) and b* (blue to yellow) are the two chromatic components, which ranges from -120 to 120. In the (L* a* b*) space, the color perception is uniform which means that the Euclidean distance between two colors corresponds approximately to the color difference perceived by the human eye (Hunt et al., 2011).

Browning depends on the frying oil temperature and on the reducing sugars content of potato strips. Accumulation of the reducing sugars in the tubers results in excessive browning of French fries and potato chips (Tran et al., 2007). Marquez and Añon (1986) investigated the influence of sugars on color development of potato strips depending on the superficial reducing sugar content. They found that fructose increased color compared to glucose. Sorbitol and xylitol as polyol's (sugar alcohols) components of coating formulations adhered to external surface of the potato samples prior frying could also influence the rate of non-enzymatic browning reactions. The type of plasticizer also demonstrated to have effect on L-values (lightness), but no consistent effect on *a*, *b*, or *ΔE* (*P* > 0.45; *P* > 0.11, *P* > 0.02, respectively; Table 4.1). Lower values of *L* represent a darker color it is expected that *L*-values decrease over frying time. This was indeed observed, with a continual and significant decrease ($P \leq 0.05$) *L* from watch frying time to the next. Formulations with sorbitol showed to have significantly increased *L*-values compared to non-coated samples indicating that the presence of the coating material resulted in lighter samples compared to non-coated potato strips. These results are in agreement with previous study carried out in potato strips (Garcia et al., 2004). Formulation MC-0.75S exhibited the highest *L*-values (54.30; Table 4.1). Furthermore, frying time and plasticizer concentration were found to be significant ($P \leq 0.05$). Improved color quality is associated with high *L*-values (Marquez & Añon, 1986).

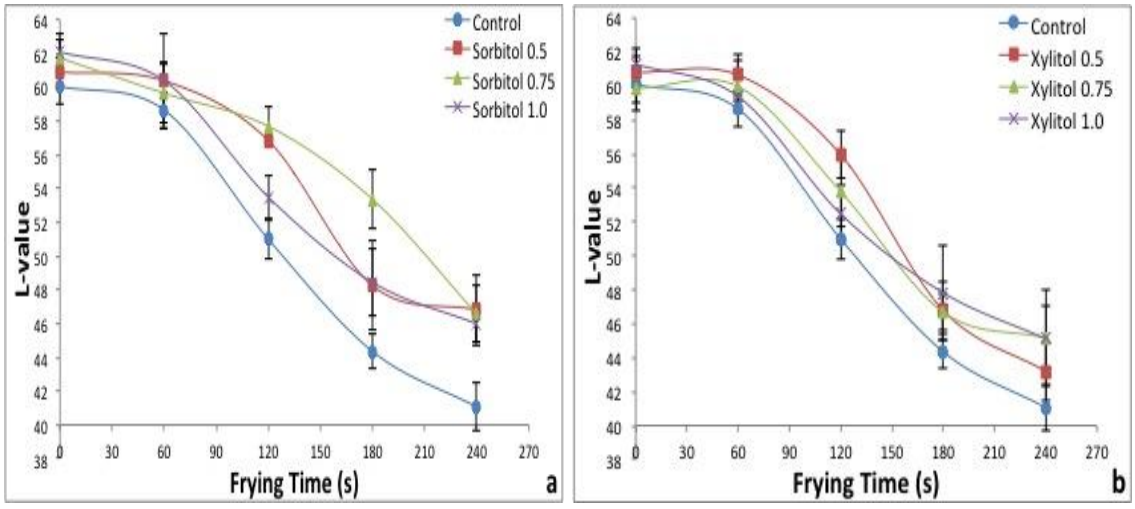

Figure 4.5. L-values in fried coated potato strips – MC sorbitol (a) and xylitol (b).

The color parameter *a** (red-green) was significantly influenced as frying time progressed. Across all coating formulations *a*-values increased from a negative value at 60 s frying, to significantly greater positive values at following frying times. The *a** values increased significantly from 60 to 120 and from 120 to 180 s frying time, but the *a** value from 180 to 240 s did not showed a significant increased value (Figure 4.6). The nature of the plasticizer, the concentration and frying time showed no significant differences between coated and non-coated samples ($P > 0.05$). In general as frying progresses the coated and non-coated potato strips shift from greener to redder color.

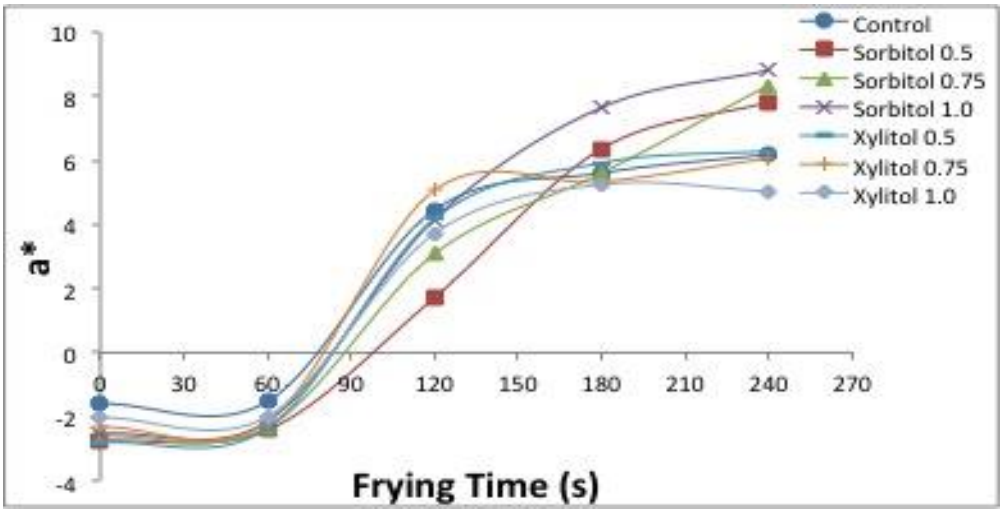

Figure 4.6. a*-values (red-green) in fried coated potato strips for both MC - sorbitol and xylitol formulations.

The color parameter *b* (yellow-blue) was not affected by the type of plasticizer, but did significantly increased over frying time ($P \leq 0.0001$; Figure 4.7), with increasing values as frying time proceeded. Potato strips showed no differences with either type of plasticizer or concentration applied in each formulation, but it did show significant differences over frying time between coated and non-coated samples. Positive values in parameter "b" (yellow-blue) are desirable in fried potatoes; *b parameter increases constantly when temperature and frying time are raised (Krokida et al., 2001).

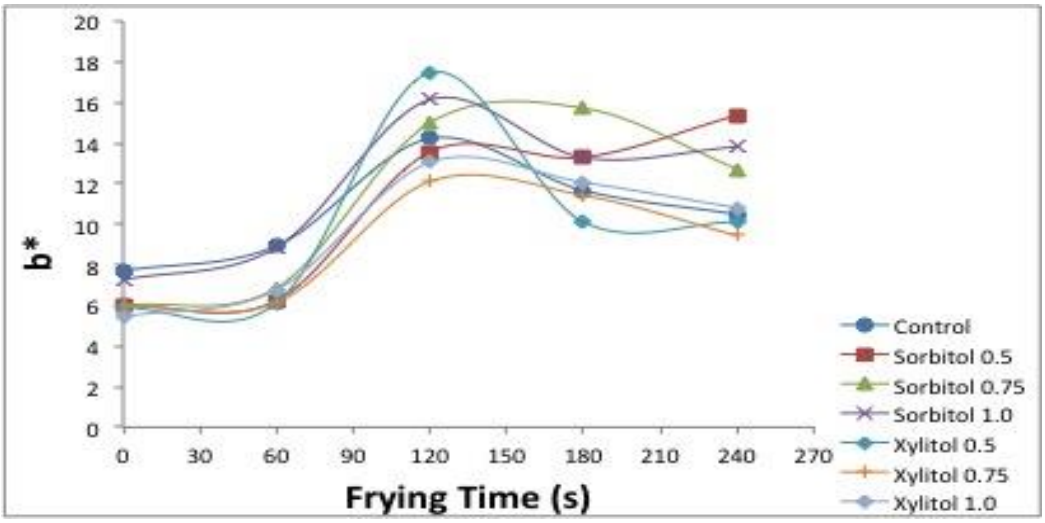

Figure 4.7. b*-values (yellow-blue) in fried coated potato strips for both MC - sorbitol and xylitol formulations**.**

In the case of *ΔE* color parameter, the type of plasticizer applied and frying time exhibited no significant differences (marginal $P < 0.14$). *ΔE* as a parameter recorded lowest values for xylitol than sorbitol potato strips. The presence of absence of the coating materials had relatively little influence in color development. Coated potato strips were observed to be lighter than non-coated potato strips. This can be explained by a reduction in the rate of drying and by the capacity of each coating to act as an external barrier reducing changes in the surfaces of potato strips.

4.4.4 Effect of coating formulations on mechanical properties of fried potato strips

The force at maximum load (F_{max}) was significantly influenced by frying time ($P < 0.0001$; Table 4.*1).* While non-coated potato strips tended to show slightly lower F_{max} values (2.83 N) than coated strips (2.96 - 3.50 N); with the exception of MC-1.0X, which resulted in even lower values than the ones recorded for NC samples. The use of sorbitol as plasticizer led to a greater $F_{max,}$ (*i.e.*, a marginally greater improvement over the non-coated samples) than the use of xylitol. In general, the use of a coating improved hardness over non-coated samples, and sorbitol contributed greater hardness than xylitol, though in both cases the differences were not significant.

The F_{max} value increased significantly from one time to the next, up to the full 240 s frying time exhibiting the highest value of 3.50 N (Figure 4.8). This can be explained by an increment in dry matter of potato strips studied related to the hardness of the material (Tajner-Czopek et al., 2008). The relative increase in F_{max} was greatest at 180 s, and was least at 60 and 240 s. At initial stage of frying potato tissue softened became cooked, and a later stage crust formation starts and progressively hardened (Pedreschi and Moyano, 2005a). The gradual increase of potato strips' F_{max} over frying time was the result of crust formation due to water loss on the external surface of fried potatoes and external crust formation (Pinthus et al., 1995).

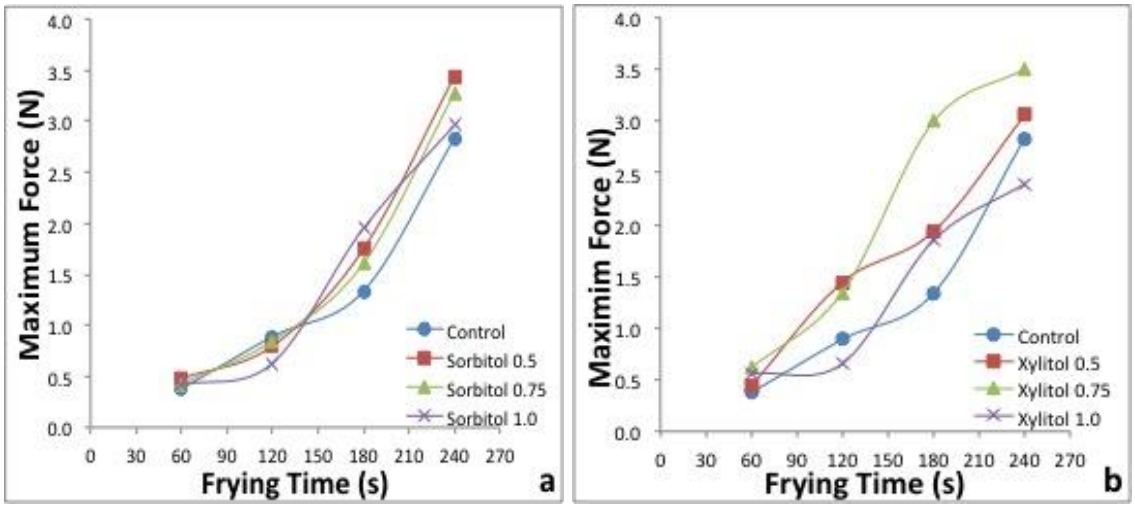

Figure 4.8. F_{max} (N) values in fried coated potato strips – MC sorbitol (a) and xylitol (b).

Young's modulus (λ), as a function of the stiffness of fried samples was also recorded. Marginally significant differences ($P \leq 0.027$) were noted for the nature of the plasticizer and concentrations analyzed (none + 6 coating formulations), but the differences (Duncan's, $P \leq 0.05$) showed that sorbitol-coating formulations increased λ values; however, the increment observed was not significant ($P > 0.05$). The value of λ increased significantly as frying progressed (0.17-3.01 N mm^{-1}). As with F_{max} the mechanical properties changed substantially over the last minute of frying (180 *vs.* 240), indicating that much of the desirable mechanical properties were developed at the end of the frying process.

4.5 Conclusions

The application of methylcellulose coatings plasticized with sorbitol (S) and xylitol (X) demonstrated to act effectively as mass transfer barriers. The nature of the plasticizer applied demonstrated to have no significant influence in moisture loss responses; however, the concentration did show significant effects on MR. MC-0.5S showed greatest MR values (0.21). Thus, they exhibit better barriers for the reduction of moisture loss rates during frying primarily at the first minutes of frying (60-120 s). Non-coated samples gained more total fat content *(%FC)* than did any of the coated potato strips. The most suitable methylcellulose coating formulation regarding the reduction of oil content in percentage was MC-0.5S (36.99%) at 240 s frying time. The oil uptake was found to be correlated with the moisture loss within coated and non-coated samples exhibiting higher coefficients of correlations ($R^2 \geq 0.93$).

The presence of the coating materials increased significantly *L*-values, thus coated potato strips exhibited improved optical properties. No significant differences were recorded on coating concentrations. Little or marginal influence on color development was recorded. Mechanical properties demonstrated to increase over frying time for both coated and non-coated samples, being more evident at last minutes of frying. Coated potato strips recorded similar (no significant differences were recorded) values than non-coated potato strips. Similarly to F_{max} values, young's modulus as a function of stiffness in coated samples did not record significant differences with respect to non-coated samples. The values of F_{max} and λ increased significantly over last minutes of frying (180-240 s) suggesting that texture development take place after 120 s of frying.

4.6 References

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CONNECTING TEXT

In chapter IV, the influence of methylcellulose coatings plasticized with sorbitol and xylitol on moisture loss, oil absorption, color development and mechanical properties were determined. The effect of frying time was significant in all the variables studied among coated and uncoated potato strips with the exception of ΔE color parameter. Coating suspensions demonstrated to deliver a product with low-fat content, enhanced optical properties, and slightly similar texture development. In chapter V, a mathematical modelling was used to correlate experimental data recorded to predicted values based on mass transfer phenomena related to first order kinetic reaction for both WPI and MC coating formulations to determine their effectiveness as coatings to improve quality in coated potato strips during deep-fat frying.

V. KINETIC MODELLING OF MOISTURE LOSS AND FAT UP-TAKE DURING DEEP FAT FRYING OF COATED POTATO STRIPS WITH DIFFERENT COATING FORMULATIONS.

5.1 Abstract

A mathematical model was used to characterize moisture loss and fat uptake in coated and non-coated fried potato strips. Two different film-forming materials were tested: isolated whey protein (WPI) and methylcellulose (MC) at 10 and 1% concentration, respectively. Two plasticizers were used to create the coating formulations [sorbitol (S) or xylitol (X)], each at three different concentrations [0.50%, 0.75% or 1.00% (w/w)]. Mass transfer (moisture loss and oil up-take) as a function of frying time was described by a first order kinetic equation. Differences in moisture loss and fat absorption were observed to be much more meaningful at the beginning of frying process. As frying progressed fat up-take decreased in intensity. The kinetic model accurately fitted the experimental values of moisture loss and oil uptake during frying in coated and non-coated potato strips, similar correlation coefficients (r^2) with respect of values predicted by the model. Mass transfer showed a strong relation with the applied film-forming materials. In contrast, the nature of the plasticizer used showed lower r^2 values, suggesting that the effect of plasticizer on mass transfer was minor. Values recorded on moisture loss and fat uptake responses for both film-forming materials fitted to the model proposed.

5.2 Introduction

Widely used in food processing, deep fat frying involves completely immersing food in hot oil medium. Frying is preferred among other cooking methods for creating unique flavors and texture in final products (Moyano & Pedreschi, 2006). Crust formation provides the most pleasant characteristics of fried products (Keller et al., 1986). Potatoes are the most important fried products from a market share point of view (Costa & Oliveira, 1999). Deep fat frying is a simultaneous heat and mass transfer operation, where heat transfer at high moisture is combined with dehydration of the food product (Gupta et al., 2000). Heat is transferred from the oil medium to the food, causing water evaporation from the food item to the surrounding medium, and the absorption of a certain amount of oil

from the medium into the food item. Heat and mass transfers are influenced by frying conditions (i.e. oil temperature, type of oil, sample geometry) as well as by the application of pre-treatments (*i.e*., air drying, osmotic drying) or by the presence of coating materials on the external surface of the food items. Thus, understanding moisture loss and fat uptake during deep fat frying is important in determining and maintaining the quality of fried foods (Ngadi et al., 1997).

Among the several authors having studied heat and mass transfer, Pravasani and Calvelo (1991) proposed that the moving boundary layer, which separates the fried potato crust and its interior, reflects the independence of strip centre and oil temperatures. Rice and Gamble (1989) noted that the driving force for the mass transfer was given by the change in the state of the water at the surface of the potato from liquid to steam, and the ensuing water loss at the potato surface. According to Fick's first law of diffusion, resistance to mass transfer is dictated by the internal resistance to mass diffusion as well as by surface resistance. Krokida et al. (2001) reported that water loss rates increased with temperature, and were greater in thinner samples. McDonough et al. (1993) observed a formation of large and small diameter holes in the surface and a network of tunnels, and cavities in the interior formed by the release of stream from the interior to the external surface of the chips. Oil was found to diffuse into tortilla chips as a result of the negative pressure in the tunnels after the steam was vented. Moreira et al. (1995) simultaneously modeled heat and mass transfer of tortilla chips during frying, using first order exponential diffusion equations for oil uptake. They demonstrated that moisture loss and oil uptake rates were faster during the first 15 s of frying, becoming fairly constant thereafter. Following the Galerkin method and using finite element equations based on governing equations of moisture transfer, Ngadi et al. (1997) proposed a mathematical heat and mass transfer model for chicken drumsticks. They demonstrated that moisture diffusivity could occur in both liquid and vapour phases. Farkas et al. (1996) predicted values for moisture loss, fat absorption and crust development in French fries based on a one-dimensional model, proposing that diffusion in the system was a function of oil temperature and followed an Arrhenius type equation. Costa and Oliveira (1999) developed a compartmented model to describe water loss in potato strips during deep-fat frying, stating that water loss was not uniform throughout the potato slice, but rather occurred

predominantly at the beginning in the edges, and as heating proceeds, gradually includes the potato core.

Models for frying and mass transfer in foods coated with edible films have been reported. Ateba and Mittal (1994) described a model for deep fat frying of beef meatballs, taking into account the influence of heat related to moisture and fat transfer. The model used general diffusion equations for heat, moisture and fat transfer assuming that foods with initially fat contents undergo two fat transfer periods: (1) fat diffusion from the surroundings into the product and (2) fat migration from the product to the surroundings by capillary flow. Based on Fourier's and Fick's laws, the Williams and Mittal (1999) model for the frying of edible coated foods considered heat and mass transfer in the film as well as in the food item itself. Most of the previous models of heat and mass transfer presented did not consider the presence of an edible film coating in the external layer of the food item. The objective of this study was to model moisture loss and fat transfer of coated potato strips, comparing the effect of two different film-forming materials: whey protein isolated (WPI) and methylcellulose (MC) based coatings, plasticized with either sorbitol (S) or xylitol (X) at three different concentrations on oil absorption and water evaporation as a function of frying time.

5.3 Model development

Driving force for mass transfer, during frying, is provided by conversion of internal water of potato strips to the outside layers in a steam phase. The surface resistance is normally limited by the rate of the phase conversion. Evidence has demonstrated that oil absorption occurs fastest at the external surface of the potato rather than the core, as oil penetrates into the system through the channels created by the release of vapour (Rice & Gamble, 1989; Costa & Oliveira, 1999).

Mass transfer was considered to occur only at the surface of the potato samples where heat transfer was most affected by evaporative cooling (Williams & Mittal, 1999). A first order kinetic model (Krokida et al., 2000), was chosen to describe moisture loss and oil up-take within the potato strip food system:

$$\boldsymbol{O} = \boldsymbol{O}_{\boldsymbol{eq}}^{\boldsymbol{1}}(\mathbf{1} - \boldsymbol{exp}\,(-\boldsymbol{K}^{\mathbf{1}}\boldsymbol{t})) \quad \textbf{(5.1)}$$

where,

t	Frying time (s)
O_{eq}^{1}	Equilibrium moisture loss and oil content, respectively (w/w).
K^1	Specific rate for a first-order model.

In the model when t = 0, oil content is null and reaches equilibrium as the frying process proceeds. A non-linear regression model was used to estimate parameters of Eq. (5.1), using SAS System software (Version 9.3, SAS Institute, Inc, 1999, Cary, NC, USA).

5.4 Materials and methods

5.4.1 Film formation

5.4.1.1 Protein-based suspensions

Preparation of protein-based coatings consisted of mixing whey protein isolated (Canada Protein, ON, Canada) with sorbitol (BP439-500; Fisher Scientific, ON, Canada) and xylitol (Fisher Scientific, ON, Canada). Aqueous suspensions of 10% whey protein isolate (WPI) were used, as films did not form below 8% whereas gels were formed above 10% (Dragich & Krochta, 2010). Sorbitol (S) and xylitol (X) plasticizer concentrations of 0.50, 0.75, and 1.00% (w/w) were tested, resulting in six coating formulations: WPI-sorbitol 0.5% (WPI-0.5S), WPI-sorbitol 0.75% (WPI-0.75S), WPI-sorbitol 1.0% (WPI-1.0S), WPI-xylitol 0.5% (WPI-0.5X), WPI-xylitol 0.75% (WPI-0.75X) and WPI-xylitol 1.0% (WPI-1.0X). The WPI (15 g) was weighed and slowly dispersed in 150 ml of distilled water under constant stirring. Solutions were heated up to 90 ± 2°C and kept at this temperature for 30 min to allow film formation. Plasticizers were incorporated into the solution after the WPI was completely dissolved.

5.4.1.2 Methylcellulose-based suspensions

Preparation of methylcellulose-based coatings consisted of mixing methylcellulose M352-500 (Fisher Scientific S.A New Jersey, USA) with sorbitol BP439-500 (Fisher Scientific, ON, Canada) and xylitol AC22598-0250 (Fisher Scientific, ON, Canada). Aqueous suspensions of 1% MC were used. Different concentration of sorbitol

(S) and xylitol (X) plasticizer (namely 0.50, 0.75, and 1.00% w/w) were tested; resulting in six coating formulations: MC-sorbitol 0.5% (MC-0.5S), MC-sorbitol 0.75% (MC-0.75S), MC-sorbitol 1.0% (MC-1.0S), MC-xylitol 0.5% (MC-0.5X), MC-xylitol 0.75% (MC-0.75X), MC-xylitol 1.0% (MC-1.0X). 1 g MC was weighed and slowly dispersed in 30 mL of hot distilled water at 100 °C under constant stirring. Once a homogeneous solution was obtained (approximately 1 h), total volume was completed to 100 mL by adding cold distilled water to the solution under stirring condition. Plasticizers were incorporated after MC was completely dissolved.

5.4.2 Sample preparation and frying medium

Potatoes (cv. 'Russet Burbank') purchased from a local supermarket (Supermarchés Provigo, Montreal, QC, Canada) were held at room temperature (~21°C). Russet Burbank potatoes were chosen due to their availability throughout the period of time the research was conducted. Tubers were manually peeled and peeler and immediately cut into slices 2.5 - 3.5 mm thick. Samples potato strips were weighed, then dipped in the coating suspensions for 20 - 40 s, weighted again, and then fried. Oil temperature was continuously monitored for each batch.

5.4.3 Experimental procedure

Prepared samples were weighed and their thickness was measured. Once coating was applied in samples they were weighted again to obtain values of film pick-up rates for each coating formulation. Samples were fried (time and temperature of frying), and excess of surface oil was removed. Samples were cooled and weighed. Moisture content was determined by gravimetric method using a forced air convection oven (Isotemp 700, Fisher Scientific, Pittsburgh, PA, USA) following AACC, 1986 official methodology. The total content *(FC)* of fried samples was determined from dried and finely ground (coffee grinder) material using the standard (AOAC, 1990) procedure (Soxhlet method). A Soxhlet extractor (SER 148, Velp Scientifica, Usmate, Italy) was used to determine fat content in samples method. Experiments were performed in triplicate.

5.5 Results and Discussion

The model was fitted to the experimental data obtained for mean moisture loss and fat absorption. Frying time had significant influence on the rate of mass transfer phenomena ($P \leq 0.05$). Experimental data fitted predicted values for both plasticizers sorbitol (S) and xylitol (X) at the three concentrations tested and for both WPI and MC. Experimental and predicted values of moisture loss rates were fitted to the proposed mathematical model for both WPI and MC film forming materials. Mass transfer phenomena - as expected - were of much greater magnitude at early frying times, and as frying progressed, the rate of oil absorption and moisture loss lower their intensity. Moisture loss responses varied according to the coating formulation applied. Non-coated samples exhibited the highest moisture loss rates in both their model-predicted and experimental values. Correlation coefficients (r^2) for the modeled *vs.* measured values were observed to be high for both film-forming materials (WPI = $0.93 < r^2 < 0.97$; MC = $0.97 < r^2 < 0.99$). Thus, it can be said that the model fitted very well with the experimental data recorded for each coating formulation analyzed on moisture loss responses. WPI-1.0X and MC-0.5S coating formulations exhibited the lowest rates of moisture loss for each film-forming material studied.

Experimental and predicted values of fat up-take were fitted to the proposed mathematical model for both film-forming materials. Data recorded on oil content O_{eq}^{1} for both WPI and MC coated samples increased as frying proceeded. Similar, to moisture loss rates, fat up-take exhibited high coefficients of correlation between observed and modelled values ($0.84 < r^2 < 0.99$ for WPI, and $0.93 < r^2 < 0.98$ MC). This suggests that the model fitted well the experimental values recorded.

Although the model fitted the experimental data overall, predicted moisture and oil equilibrium values were lower than experimental values at the last frying time. Moyano and Pedreschi (2006) also recorded this behaviour using first order kinetic equation.

The study demonstrated that equilibrium oil content (O_{eq}^{1}) differed for each coating suspension, where the highest value was found for non-coated samples. The rates of moisture loss observed correlated with the values of total fat up-take; the highest moistures loss rates resulted in higher rates in fat up-take and vice versa. The lowest value of equilibrium oil content (O_{eq}^{1}) in WPI coatings was recorded in WPI-1.0% coating

formulation, while for MC coatings suspensions, the MC-0.5S formulation had the same effect., Since both experiments were conducted separately, oil up-take absorbed in each coating formulation applied in potato strips was compared to the value given by each non-coated group rate.

Variations in mass transfer phenomena observed could be attributed to the effectiveness of each coating formulation in the reduction of moisture loss and fat diffusivity. Thus, the coated material tested indeed acted as a barrier reducing mass transfer phenomena.

Film forming materials had greatest effects on moisture loss recording higher r^2 (predicted *vs.* measured) values [(WPI-1.0X, r^2 = 0.95; MC-0.5S, r^2 = 0.98). Comparatively, when the data was fitted to account for the effect of the nature of the plasticizer, the correlation was high for sorbitol formulations (r^2 = 0.80), but poor for xylitol coatings (r^2 = 0.52). In the case where plasticizers applied were compared, the highest correlation in moisture loss responses were observed for WP-xylitol (r^2 =0.97) and MC-sorbitol (r^2 = 0.99)] (Figure 5.1).

Similar to moisture loss responses, fat up-take rates recorded a direct relationship with the film-forming material applied in the coating formulation. MC coatings exhibited higher coefficients of correlation (r^2 = 0.96), than WPI suspensions (r^2 = 0.93). The nature of the plasticizer applied again exhibited lower r^2 values (sorbitol = 0.36; xylitol = 0.10), suggesting that the effect of the nature of the plasticizer was not important in the model proposed. When the film forming material and the plasticizer was taken in consideration the r^2 values were higher for WPI-xylitol (r^2 = 0.97) and MC-sorbitol (r^2 = 0.96) formulations (Figure 5.2).

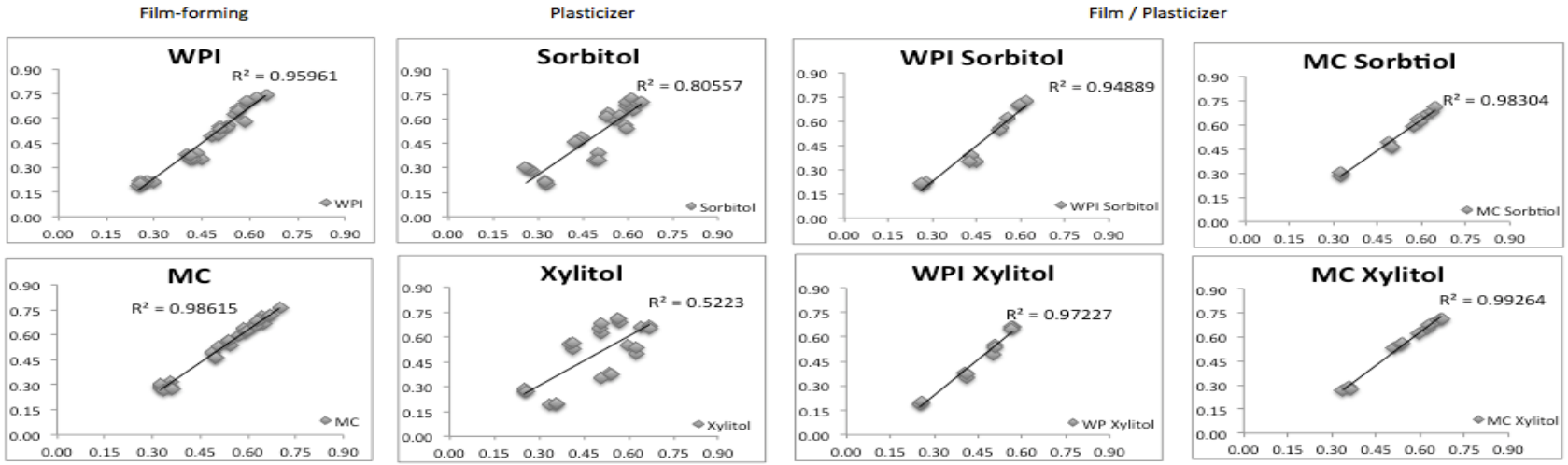

Figure 5.1. Data fit in proposed mathematical model for moisture loss responses for both WPI and MC coating formulations.

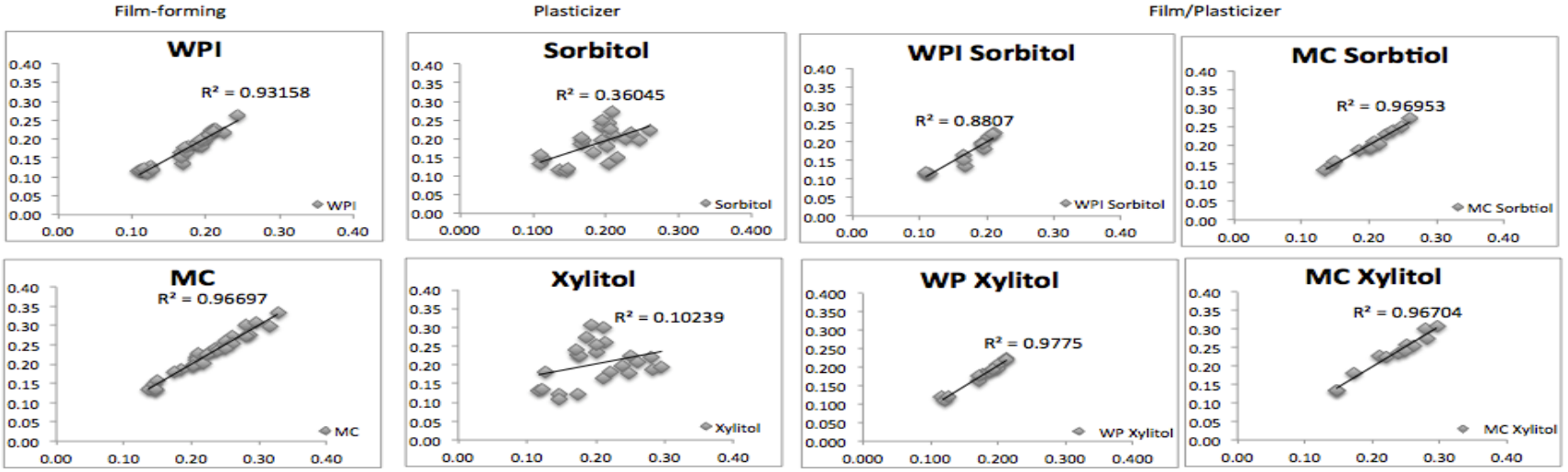

Figure 5.2. Data fit in proposed mathematical model for total oil up-take responses for both WPI and MC coating formulations.

5.6 Conclusion

Moisture loss and oil uptake were directly dependent to frying time. Being much more evident in uncoated samples. As expected, moisture loss rates and total fat up-take varied upon each coating material tested giving higher r^2 values than the ones observed when considering just the nature of the plasticizer applied. The study demonstrated that mass transfer values fitted predicted and experimental values validating the model proposed. The highest coefficients of correlation (r^2) were observed when film-forming material data was grouped with the type of plasticizer applied to create each coating formulation. All coated potato strips decreased the rates of mass transfer suggesting that the formulation in both film-forming materials was effective to deliver potato strips with reduced mass transfer rates.

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VI. GENERAL SUMMARY

6.1 General conclusions

During the past years the health concern in fat consumption has led to the need of studying novel products and procedure to successfully reduce fat absorption in fried products. Research conducted previously has demonstrated the effectiveness of whey protein isolated (WPI) and methylcellulose (MC) as hydrocolloid materials in the formation of coatings to act as barriers controlling fat absorption during frying process. Moreover, the application of both coating materials: whey protein isolate and methylcellulose hydrocolloids contributed not only to the nutritional point of view by delivering low-fat products, but also creating products with similar color development and textural properties.

WPI-coatings showed a constant increase in the initial moisture content by 0.50% compared to non-coated samples due the presence of the coating material in the external surface of the potato strips analyzed. The relative decrease in moisture loss was observed to be greater in coated samples suggesting that the coating film has proved to improve water evaporation. Fat content was recorded to be significantly higher in non-coated samples. Moreover, WPI-1.0X coating formulation exhibited the greatest reduction rates in mass transfer, exhibiting greatest values of moisture content and lowest values of total reduction in fat uptake (*MC = 18%; FC =27%*) at 240 s of frying time. Color development demonstrated to begin as frying proceeded. The presence of the coating materials increased significantly *L*-values, thus coated potato strips exhibited improved optical properties. Mechanical properties demonstrated that coated samples had significantly increased force at maximum load (F_{max}) than that non-coated potato strips exhibited. The use of sorbitol as a plasticizer in coating formulations led to greater F_{max} values, improving hardness over non-coated samples. Marginally significant differences were noted on Young's modulus (λ) as a function of stiffness between coated and non-coated samples.

MC-coatings exhibited a steady increase of 0.45% in the initial moisture content compared to non-coated samples. Moisture loss demonstrated to be significantly lower in coating formulations. Lower concentrations in the plasticizer (0.5%) applied led to greater relative increase in moisture content rates suggesting that water evaporation was reduced

during frying time, especially during the first minutes of frying (60-120 s). Fat content was significantly higher in non-coated samples. Moreover, MC-0.5S coating formulation showed to be the most suitable coating formulation for the reduction of mass transfer, recording highest values of both moisture content and relative reduction of fat content (MC = 17.83%; *FC* = 36.99%) at 240 s of frying time. The presence of coating formulations increased significantly *L*-values (lightness), suggesting that coated potato strips have improved optical properties compared to non-coated samples. In overall, the presence of the coating materials demonstrated to have marginal influence on color parameters (a*; b*). Mechanical properties exhibited no significant differences among coated and non-coated samples in both F_{max} and Young's modulus parameters. Textural properties demonstrated to increase significantly at last minutes of frying (180-240 s).

The mathematical modelling proposed using first order kinetic equations correlate a good fit between experimental and predicted data for both WPI and MC film-forming materials in mass transfer phenomena (moisture loss and fat-uptake responses) during deep-fat frying. Mass transfer (moisture loss and oil up-take) as a function of frying time was described by a first order kinetic equation. Moisture loss and fat absorption were observed to be much more meaningful at first stage of frying (60-120 s). Experimental and predicted values fitted to the model proposed. Film-forming materials had strong connection to the responses of both moisture loss and fat-content. Contrary, when data was grouped by the nature of the plasticizer applied this relation was poorer giving lower values of coefficient of correlation, suggesting that the nature of the hydrocolloid material applied has greater relation in mass transfer responses. The highest coefficients of correlation (r^2) were observed when film-forming material data was grouped with the type of plasticizer applied to create each coating formulation. All coated potato strips showed to have decreased the rates of mass transfer suggesting that the formulation in both film-forming materials was effective to deliver potato strips with lower fat up-take.

When both film forming materials (WPI & MC) were compared to determine their overall functionality as coatings to improve quality in fried coated potato strips. MC-based coatings were more effective in terms of: total fat reduction (FC) 36.99 *vs*. 27%, respectively; improved optical properties by recording higher *L*-values than WPI coated

samples (54.30 vs. 53.47); increased mechanical properties than those recorded by WPI coated samples [F_{max} (3.50 *vs.* 2.98); Young's modulus (3.01 *vs.* 2.98), respectively]

6.2 Recommendations for future research

Further investigation has to be conducted in order to fully understand the complex relation between the type of film-forming material (hydrocolloid) and the nature of plasticizer when developing new coating formulations. The concentration of the plasticizer applied demonstrated to play a crucial role in the behaviour and overall effectiveness of the film to act as a barrier decreasing mass transfer. This would provide the knowledge to create enhanced films with improved properties resulting in low-fat fried products.

Since mechanical and external appearance of food items are the most important parameters of consumers' evaluation of quality in a food product. The need to understand the role of each film forming material in the external surface of coated fried samples would help choosing appropriate hydrocolloid material according to the physicochemical properties of the substrate applied. Coated samples showed a slight improve in the overall optical properties. Therefore, the need to understand changes in Maillard reactions and a possible increase in sugars coming from the addition of plasticizers needs to be assessed. Mechanical properties marginally varied depending upon the nature of the plasticizer applied in the coating formulation. Thus, the necessity to understand the role of the plasticizer and its concentration is evident.

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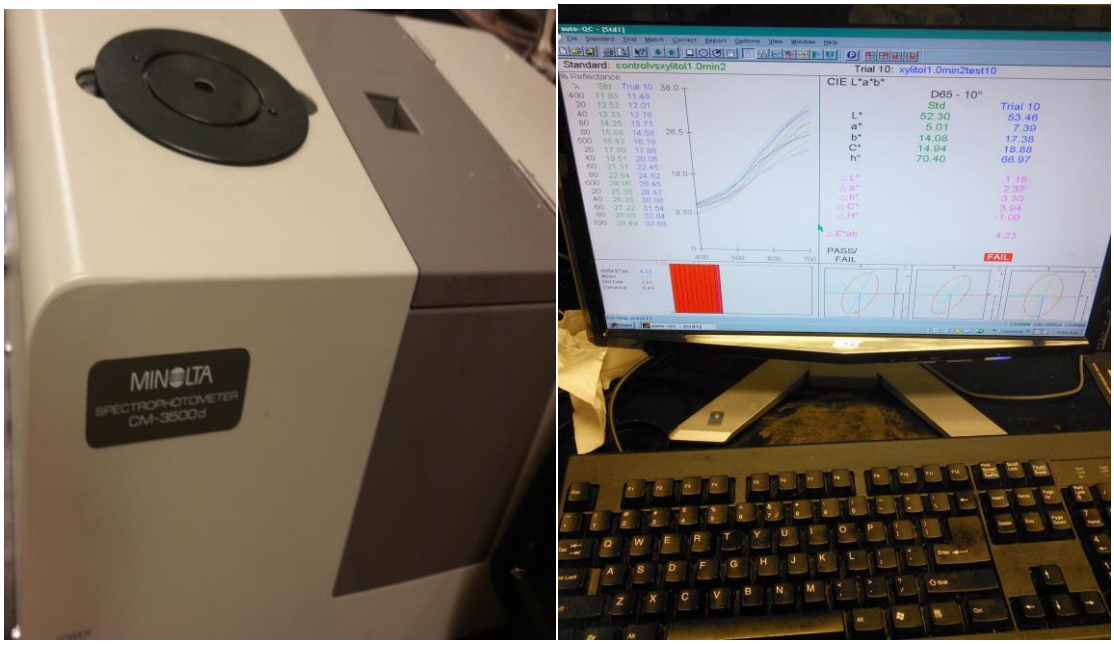
APPENDICES

APPENDIX 1.1: Soxhlet extractor - Velp Scientifica**.**

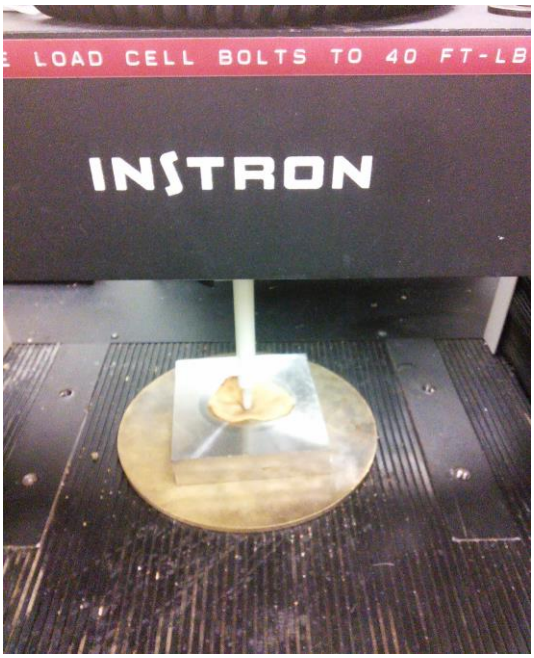
The extractor Soxhlet SER 148 (Velp Scientifica, Usmate, Italy) used in this study is shown above to determine fat content in samples previously ground. Fat was extracted using petroleum ether as solvent. The equipment is located at the department of Bioresource Engineering, Macdonald Campus of McGill University.

APPENDIX 1.2: Konica Minolta colorimeter



The colorimeter Konica Minolta (CR-300, Konica Minolta, Sensing, Inc. Osaka, Japan) used in this study to determine the color intensity of samples previously calibrated with a standard white ceramic plate. The equipment is located at the department of Bioresource Engineering, Macdonald Campus of McGill University.

APPENDIX 1.3: Instron Universal Testing Machine



Instron Universal Testing Machine (Model 4502, Canton, MA, USA) used in this study to determine mechanical properties of samples by compression tests. The equipment included a 5 mm diameter probe.

APPENDIX 2.1: Whey protein isolated nutritional value.

CANADIAN PROTEIN.COM

100% PREMIUM WHEY ISOLATE

0.9kg (2lbs) 2.27kg (5lbs) 11.34kg (25lbs) 20.41kg (45lbs)

Nutrition Facts
Valeur nutritive

Per 30 g (1 scoop) / par 30 g
Servings Per Pound: 15

Amount / Teneur	%DV / %VQ
Calories / Calories 122	
Fat / Lipides 0 g	0 %
Saturated / saturés 0 g	0 %
+ Trans / trans 0 g	
Cholesterol / Cholestérol 0 mg	
Sodium / Sodium 0 mg	0 %
Carbohydrate / Glucides <0.5 g	0 %
Fibre / Fibres 0 g	0 %
Sugars / Sucres 0 g	
Protein / Protéines 29 g	61 %
Vitamin A / Vitamine A	2 %
Vitamin C / Vitamine C	10 %
Calcium / Calcium	38 %
Iron / Fer	0 %

*%Daily Values based on 2000 calorie diet.

Amino Acid profile

Amount Per 100 grams

Alanine	4.7 g
Arginine	2.3 g
Aspartic Acid	11.1 g
Cystine	3.0 g
Glutamic Acid	16.5 g
Glycine	1.6 g
Histidine	2.2 g
Isoleucine	5.4 g
Leucine	11.9 g
Lysine	11.1 g
Methionine	2.3 g
Phenylalanine	3.5 g
Proline	4.4 g
Serine	2.7 g
Threonine	4.4 g
Tryptophan	2.7 g
Tyrosine	3.5 g
Valine	5.5 g

Directions: As a dietary supplement, mix 1-2 servings of material in approximately 12-16 fl oz of your favorite beverage. Stir, and enjoy!

Caution: Not intended for children. Consult a healthcare practitioner prior to use if you are pregnant or breastfeeding. Consult a healthcare practitioner prior to use if you have health conditions. Store in a cool dry place away from children. Do not use if seal is broken.

Allergy Statement: Contains: Milk, and traces of Soy Lecithin for instantizing. Manufactured in a facility that also processes eggs, peanuts, wheat, tree nuts, sulphites, sesame and seafood.

Manufactured for CanadianProtein.com - Ontario, Canada
For more information visit: www.CanadianProtein.com
Made in Canada

Whey protein isolate (Canada Protein, ON, Canada) used to prepare protein-based coating formulations analyzed in this study.

APPENDIX 2.2: Methylcellulose specifications.

Product Specifications	
Assay	27.5 to 31.5% (FCC/USP)
Lead [Pb]	3mg/kg max. (FCC)
Viscosity	3000 to 5600 CPS (FCC/USP)
Heavy Metals [as Pb]	10mg/kg max. (FCC), 0.001% max. (USP)
Identification	Pass Test (FCC/USP)
Loss on Drying	5.0% max. (FCC/USP)
Ignition Residue	1.5% max. (FCC/USP)
Residual Solvents	Meets Requirements (USP)
Additional Information	Vapor Pressure: Negligible

Methylcellulose M352-500 (Fisher Scientific S.A New Jersey, USA), used to prepare polysaccharide-based coating formulations.

APPENDIX 3.1: ANOVA results for the different parameters analyzed in protein-based coatings.

Parameter	Model and Factor Significance (Type III SS)								
	MODEL			COATING			FRYING TIME		
	d.f.	*F*	*P***	d.f.	*F*	*P*	d.f.	*F*	*P*
*%MC**	10	72.91	**<0.0001**	6	3.56	**0.016**	4	176.95	**<0.0001**
%FC	9	30.84	**<0.0001**	6	3.15	**0.027**	3	86.21	**<0.0001**
L	9	18.96	**<0.0001**	6	1.88	0.141	3	129.25	<**0.0001**
a*	9	3.17	**0.0180**	6	1.76	0.165	3	6.00	**0.005**
b	9	6.85	**0.0003**	6	1.12	0.391	3	18.30	**<0.0001**
ΔE	9	8.15	**0.0080**	6	1.04	0.355	3	29.22	**<0.0006**
F_{max}*	9	20.59	**<0.0001**	6	4.25	**0.008**	3	53.26	**<0.0001**
λ	9	15.79	**<0.0001**	6	2.37	0.073	3	42.64	**<0.0001**

*for transformed data, ** *P* values in bold show significant differences at α = 0.05

Parameter	Model and Factor Significance (Type III SS)													
	MODEL		PLAST		[PLAST]		TIME		PLAST* [PLAST]		PLAST *TIME		[PLAST] *TIME	
	F	*P*	*F*	*P*	*F*	*P*	*F*	*P*	*F*	*P*	*F*	*P*	*F*	*P*
%MLD	23.28	**0.0004**	31.69	**0.0013**	1.58	0.2804	104.1	**<0.0001**	2.28	0.1839	5.15	0.0524	4.72	0.0504
%FCD	1.15	0.4613	4.93	0.0681	0.41	0.6791	1.86	0.2368	0.23	0.8015	1.49	0.3088	0.56	0.7534
L	13.69	**0.0019**	0.78	0.4122	10.4	**0.0112**	51.84	**0.0001**	0.23	0.8012	0.73	0.5687	8.84	**0.009**
a_*	0.29	0.9787	0.06	0.8107	0.1	0.902	0.07	0.9736	0.69	0.5375	0.28	0.8371	0.38	0.8689
b_*	1.18	0.4469	1.16	0.3228	0.15	0.861	2.18	0.1918	2.89	0.1323	0.07	0.9762	1.03	0.488
ΔE	8.15	**0.0080**	1.00	0.3548	7.93	0.0207	29.22	**0.0006**	3.43	0.1017	1.36	0.3416	3.85	0.0628
F_{max}	1.89	0.2211	4.66	0.0742	1.43	0.3103	5.00	**0.0452**	2.79	0.1388	0.65	0.6117	0.35	0.8877
λ	2.88	0.098	4.06	0.0904	3.22	0.1124	6.35	**0.0272**	3.97	0.0798	0.55	0.6682	1.65	0.2798

Degrees of freedom for MODEL, PLAST, [PLAST], TIME, PLAST*[PLAST], PLAST*TIME, [PLAST]*TIME are 17, 1, 2, 3, 2, 3, 6, respectively.
**P* values in bold show significant differences at α=0.05.

APPENDIX 3.2: Mean values separated by Duncan's Multiple Range Test (DMRT)

Parameter	TREATMENT FACTOR											
	COATING							FRYING TIME (s)				
	None	Sorbitol (%)			Xylitol (%)							
		0.5	0.75	1	0.5	0.75	1	0	60	120	180	240
*MC**	40.69b	45.11ab	46.07a	46.91a	48.65a	49.02a	48.72a	84.50a	62.18b	44.46c	27.46d	13.66e
%*ML*	52.72a	47.58b	46.13b	44.98b	43.53b	43.07b	43.44b	—	20.68d	38.40c	55.40b	69.20a
%*FC*	20.07a	17.14b	16.54b	17.14b	17.73b	17.97b	17.07b	—	11.83d	16.53c	19.80b	22.49a
L	52.67b	54.63ab	53.57ab	54.92a	54.79ab	53.63ab	55.43a	—	59.60a	55.13b	52.30c	49.53d
a*	4.07ab	3.02ab	2.46b	4.49a	4.42ab	4.00ab	2.55b	—	-1.22c	4.04b	6.09a	5.37ab
b	12.86a	13.19a	13.77a	16.33a	14.44a	13.55a	13.79a	—	9.01c	15.44ab	17.11a	14.39b
F_{max}*	1.44b	1.92b	2.98a	1.75b	1.86b	1.75b	1.87b	—	0.69d	1.09c	1.90b	4.07a
λ	0.75b	1.10ab	1.15ab	1.75a	1.30ab	0.94b	0.73b	—	0.41b	0.44b	0.76b	2.79a

Parameter	TREATMENT FACTOR								
	PLASTICIZER		[PLASTICIZER] (%)			FRYING TIME (s)			
	Sorbitol	Xylitol	0.5	0.75	1	60	120	180	240
%FP	4.82a	3.40a	5.16a	3.10a	4.08a	—	—	—	—
%*ML*	13.51b	18.99a	15.03a	16.78a	16.94a	17.71b	29.65a	10.16c	7.48c
%*FC*	14.88a	9.52b	12.23a	13.53a	10.85a	8.56a	11.58a	12.12a	16.55a
L	1.04a	1.05a	1.05a	1.03b	1.06a	1.00d	1.03c	1.06b	1.09a
a*	0.85a	0.89a	0.87a	0.82a	0.92a	0.91a	0.89a	0.87a	0.81a
b*	1.12a	1.04a	1.08a	1.05a	1.10a	0.94a	1.05a	1.13a	1.20a
ΔE	3.94a	3.53a	2.87b	3.49b	4.85a	1.41c	2.77cb	4.06b	6.69a
F	2.07a	1.32b	1.48a	2.11a	1.49a	1.10b	2.14ab	2.57a	0.97b
λ	2.32a	1.43a	1.54a	1.42a	2.67a	0.87b	1.80b	3.45a	1.38b

*values are significantly different row-wise within treatment factor if their letters differ (Duncan's multiple range test, α=0.05). Shaded values are not significant within their treatment factor

APPENDIX 3.3: Test of normality for the different parameters analyzed in protein-based coatings.

Parameter	Factors	Normality test statistics*							
		W		*D*		W^2		A^2	
		Value	*P*	Value	*P*	Value	*P*	Value	*P*
MC	TREAT (7) TIME	0.934	**0.036**	0.147	0.053	0.124	**0.05**	0.809	**0.034**
ln(MC)		0.949	0.104	0.12	>0.15	0.082	0.195	0.554	0.146
L	TREAT (7) TIME (4)	0.960	0.356	0.113	>0.15	0.044	>0.25	0.31	>0.25
a		0.851	**0.001**	0.129	>0.15	0.122	0.055	0.901	**0.02**
exp(a+2)		0.937	0.093	0.117	>0.15	0.069	>0.25	0.505	0.195
b		0.959	0.325	0.103	>0.15	0.046	>0.25	0.336	>0.25
F_{max}		0.936	0.086	0.176	**0.024**	0.14	**0.031**	0.766	**0.043**
$ln(F_{max})$		0.978	0.806	0.09	>0.15	0.034	>0.25	0.217	>0.25
λ		0.979	0.818	0.116	>0.15	0.033	>0.25	0.239	>0.25
$\%MLD_{c/nc}$	PLAST (2) [PLAST] (3) TIME (4)	0.948	0.252	0.111	>0.15	0.091	0.144	0.521	0.174
$\%FCD_{c/nc}$		0.981	0.876	0.136	>0.15	0.05	>0.25	0.269	>0.25
$L_{c/nc}$		0.979	0.874	0.081	>0.15	0.042	>0.25	0.268	>0.25
$a_{c/nc}$		0.984	0.966	0.061	>0.15	0.015	>0.25	0.119	>0.25
$b_{c/nc}$		0.975	0.807	0.129	>0.15	0.06	>0.25	0.338	>0.25
ΔE		0.971	0.696	0.102	>0.15	0.033	>0.25	0.219	>0.25
$F_{max, c/nc}$		0.974	0.785	0.119	>0.15	0.05	>0.25	0.272	>0.25
$\lambda_{c/nc}$		0.977	0.83	0.087	>0.15	0.037	>0.25	0.271	>0.25
FP		0.943	0.6826	0.197	>0.15	0.042	>0.25	—	—

* Shapiro-Wilks *W*, Kolmogorov-Smirnov *D*, Cramer-von Mises W^2, and Anderson-Darling A^2. *P* values in bold show significant differences at α=0.05, indicating that the distribution of the residuals is non-normal.

APPENDIX 3.4: Mean values recorded in protein-based coatings.

Parameter		Frying Time (s)	Coating Type					
			10% w/w Whey protein (WPI)					
			Sorbitol (% w/w)			Xylitol (% w/w)		
			0.5	0.75	1.00	0.5	0.75	1.00
Moisture content	Final content (% w/w)	0	83.17	82.94	83.23	83.22	83.00	83.32
		60	61.25	62.74	61.63	64.01	64.04	63.13
		120	48.35	44.15	48.24	47.94	45.06	46.09
		180	20.83	26.82	29.02	28.41	33.46	30.01
		240	10.68	13.23	12.65	16.95	16.61	18.46
Film pick-up rate	(% w/w) per sample	60	7.85	3.49	4.17	3.32	2.80	4.11
		120	6.49	3.37	4.96	3.10	3.11	3.30
		180	6.58	2.56	4.04	4.46	3.05	3.70
		240	6.24	3.62	4.51	3.24	2.79	3.84
Fat	Final fat content (g w/w)	60	0.115	0.114	0.119	0.121	0.110	0.120
		120	0.164	0.135	0.150	0.165	0.181	0.179
		180	0.180	0.198	0.194	0.198	0.207	0.188
		240	0.226	0.214	0.222	0.225	0.220	0.196
	Fat reduction (WPI-NC)	60	9.55	10.72	6.53	6.19	14.61	6.82
		120	10.79	26.61	18.34	10.91	2.22	3.35
		180	18.60	10.20	12.06	10.53	6.34	15.13
		240	16.33	20.85	18.03	16.86	18.66	27.72
Color	L*	0	59.66	59.55	61.09	59.79	61.01	61.36
		60	58.38	59.18	60.01	59.62	61.15	59.02
		120	57.15	54.47	55.18	56.56	53.51	55.21
		180	51.92	52.86	52.70	52.32	52.69	54.03
		240	50.00	47.78	51.79	50.67	47.16	53.48
	a*	0	-1.58	-1.10	-1.14	-0.95	-1.19	-1.92
		60	-1.49	-1.05	-1.14	-0.56	-1.15	-1.82
		120	2.19	4.09	4.57	5.40	4.99	2.58
		180	5.47	6.64	7.58	5.86	5.71	4.51
		240	5.89	2.97	6.94	6.98	6.46	4.91
	b*	0	7.62	7.48	8.35	7.65	7.98	7.83
		60	8.72	10.00	9.86	9.46	9.31	6.23
		120	12.86	16.87	17.60	17.31	14.68	13.94
		180	15.65	19.36	19.06	16.40	16.62	17.23
		240	15.56	8.84	18.78	14.58	13.59	17.75
	ΔE	0	0.43	0.77	1.28	0.72	0.50	1.32
		60	4.41	1.78	1.99	2.55	2.68	3.50
		120	3.76	3.30	3.93	5.32	2.69	3.79
		180	3.84	4.02	3.93	3.90	4.44	7.29
		240	7.67	6.23	8.28	6.57	3.13	8.27
Texture	Load at maximum force (N)	60	0.71	0.94	0.78	0.63	0.47	0.61
		120	1.06	0.97	1.11	0.79	0.76	0.97
		180	1.38	2.37	2.09	1.91	1.01	1.80
		240	4.52	4.30	3.01	4.09	4.74	4.09
	Young's Modulus (N mm-1)	60	0.29	0.45	0.40	0.31	0.27	0.29
		120	0.41	0.27	0.31	0.30	0.30	0.42
		180	0.37	0.81	0.57	0.84	0.35	0.73
		240	3.33	3.06	3.58	3.76	2.85	1.47

Whey protein isolate coating formulations mean values for all parameters studied in this study after 240 s of frying time: Moisture loss, film pick-up rate, total fat content, color (L, ΔE), and texture development [Load at maximum force (N); Young's Modulus (λ)].

APPENDIX 4.1: ANOVA results for the different parameters analyzed in polysaccharide-based coatings

Parameter	Model and Factor Significance (Type III SS)								
	MODEL			COATING			FRYING TIME		
	d.f.	*F*	*P***	d.f.	*F*	*P*	d.f.	*F*	*P*
MC	10	419.02	**<0.0001**	6	3.71	**0.0095**	4	1041.9	**<0.0001**
*ML**	9	134.7	**<0.0001**	6	3.98	**0.0104**	4	396.15	**<0.0001**
FC	9	44.46	**<0.0001**	6	21.7	**<0.0001**	3	90.00	**<0.0001**
L	9	52.74	**<0.0001**	6	5.52	**0.0021**	3	147.19	**<0.0001**
a	9	39.90	**<0.0001**	6	0.99	0.4594	3	117.71	**<0.0001**
b*	9	10.15	**<0.0001**	6	2.02	0.1163	3	26.40	**<0.0001**
ΔE	9	2.09	0.1853	6	9.77	0.0204	3	2.75	0.1345
F_{max}	9	6.17	**0.0005**	6	2.19	0.0927	3	14.12	**<0.0001**
λ	9	37.48	**<0.0001**	6	3.14	0.0277	3	106.16	**<0.0001**

*for transformed data, ** *P* values in bold show significant differences at $\alpha = 0.05$

Parameter	Model and Factor Significance (Type III SS)													
	MODEL		PLAST		[PLAST]		TIME		PLAST*[PLAST]		PLAST*TIME		[PLAST]*TIME	
	F	*P*	*F*	*P*	*F*	*P*	*F*	*P*	*F*	*P*	*F*	*P*	*F*	*P*
%ML	18.5	**0.0008**	0.06	0.8156	4.43	0.0658	85.08	**<0.0001**	5.38	0.0459	5.34	0.0395	3.92	0.0604
%FC	1.61	0.2888	2.56	0.1607	0.11	0.8945	4.10	0.0668	0.58	0.5875	2.36	0.1711	0.68	0.6744
L	12.22	**0.0027**	1.05	0.3461	9.67	0.0133	45.19	**0.0002**	0.33	0.7319	0.70	0.5850	8.17	0.0109
a	0.32	0.9714	0.11	0.7473	0.16	0.8528	0.04	0.9875	0.62	0.5692	0.36	0.7814	0.41	0.8473
b	2.29	0.1559	0.40	0.5491	1.16	0.3755	5.12	0.0431	1.93	0.2249	0.49	0.7037	2.58	0.1365
ΔE	2.09	0.1853	9.77	0.0204	1.52	0.2931	2.75	0.1345	0.83	0.4803	2.07	0.2064	1.09	0.4585
F_{max}	1.99	0.2028	4.32	0.0829	1.39	0.3207	5.00	0.0452	3.35	0.1057	0.97	0.4643	0.34	0.8901
λ	2.69	0.1135	3.62	0.1058	3.30	0.1081	5.96	0.0312	3.55	0.0962	0.58	0.6499	1.45	0.3303

Degrees of freedom for MODEL, PLAST, [PLAST], TIME, PLAST*[PLAST], PLAST*TIME, [PLAST]*TIME are 17, 1, 2, 3, 2, 3, 6, respectively

**P* values in bold show significant differences at α=0.05

APPENDIX 4.2. Mean values separated by Duncan's Multiple Range Test (DMRT).

Parameter	TREATMENT FACTOR											
	COATING							FRYING TIME (s)				
	None	Sorbitol (%)			Xylitol (%)							
		0.5	0.75	1	0.5	0.75	1	0	60	120	180	240
MC	37.22c	42.89a	41.64ab	41.46ab	41.08ab	39.07bc	38.54bc	83.17a	54.14b	31.71c	19.39d	12.94e
ML	57.04a	50.51c	51.88bc	52.2bc	52.67bc	55.27ab	55.79ab	-	29.03d	51.45c	63.78b	70.23a
FC	28.18a	18.193e	20.372d	22.024cd	21.319cd	22.877c	25.05b	-	15.78d	22.21c	24.79b	27.50a
L	48.75bc	53.05a	54.30a	52.06ab	51.61b	51.39b	51.18b	-	59.85a	54.44b	47.93c	44.83d
a	3.675a	3.367a	3.672a	4.615a	3.505a	3.570a	3.000a	-	-2.127	3.764b	5.952a	6.927a
b*	11.01a	12.11ab	12.568a	13.018a	10.94ab	10.66ab	9.77ab	-	7.124c	11.817b	12.501b	14.501a
F_{max}	0.88ab	0.722b	0.712b	0.727b	1.22ab	1.572ab	1.097ab	-	0.238c	0.895b	1.074b	1.755a
λ	1.355b	1.485b	1.532b	1.607b	1.715ab	2.110a	1.360b	-	0.468d	0.938c	1.911b	3.061a

Parameter	TREATMENT FACTOR								
	PLASTICIZER		[PLASTICIZER] (%)			FRYING TIME (s)			
	Sorbitol	Xylitol	0.5	0.75	1	60	120	180	240
FP%	2.713a	2.503a	3.270a	2.370a	2.185a	-	-	-	-
%ML	11.786a	12.040a	9.746b	12.68ab	13.30a	9.846b	25.92a	4.246c	7.63bc
%FC	14.885a	11.122a	12.229a	13.535a	13.248a	8.564b	11.57b	12.12ab	19.75a
L	1.042a	1.048a	1.0479a	1.0271b	1.0611a	0.995d	1.028c	1.063b	1.094a
a	0.8493a	0.9064a	0.8717a	0.8219a	0.9400a	0.917a	0.888a	0.871a	0.834a
b	1.1173a	1.0773a	1.0764a	1.0518a	1.1637a	0.938b	1.046b	1.126ab	1.277a
ΔE	5.3080a	3.3232b	4.5393a	4.8525a	3.5550a	2.782b	4.62ab	4.671ab	5.18a
$F_{max,}$	2.0716a	1.3792a	1.4795a	2.1124a	1.5842a	1.103b	2.13ab	2.567a	1.094a
λ	2.3235a	1.4579a	1.5431	1.4154a	2.7136a	0.865b	1.801b	3.453b	1.442a

*values are significantly different row-wise within treatment factor if their letters differ (Duncan's multiple range test, α=0.05). Shaded values are not significant within their treatment factor.

APPENDIX 4.3: Test of normality for the different parameters analyzed in polysaccharide-based coatings

Parameter	Factors	Normality test statistics*							
		W		*D*		W^2		A^2	
		Value	*P*	Value	*P*	Value	*P*	Value	*P*
ML	TREAT (7) TIME (5)	0.9258	**0.048**	0.149	**0.107**	0.117	**0.063**	0.745	**0.046**
ln(ML)		0.961	0.368	0.093	>0.15	0.035	>0.25	0.295	>0.25
FC (db)		0.985	0.957	0.091	>0.15	0.030	>0.25	0.191	>0.25
L	TREAT (7) TIME (4)	0.980	0.861	0.094	>0.15	0.046	>0.25	0.290	>0.25
a		0.973	0.667	0.136	>0.15	0.050	>0.25	0.296	>0.25
b		0.905	**0.015**	0.148	**0.113**	0.141	0.029	0.898	**0.020**
ln(b)		0.889	0.006	0.160	0.065	0.166	0.014	1.019	0.009
F_{max}		0.961	0.368	0.136	>0.15	0.087	0.159	0.504	0.195
λ		0.980	0.858	0.119	>0.15	0.041	>0.25	0.233	>0.25
ML	PLAST (2) [PLAST] (3) TIME (4)	0.923	0.071	0.149	>0.15	0.109	0.081	0.230	0.067
FC		0.981	0.921	0.119	>0.15	0.056	>0.25	0.290	>0.25
L		0.975	0.796	0.124	>0.15	0.062	>0.25	0.339	>0.25
a		0.985	0.975	0.064	>0.15	0.015	>0.25	0.108	>0.25
B		0.975	0.795	0.101	>0.15	0.028	>0.25	0.187	>0.25
ΔE		0.987	0.986	0.052	>0.15	0.011	>0.25	0.096	>0.25
$F_{max,}$		0.975	0.793	0.121	>0.15	0.046	>0.25	0.261	>0.25
λ		0.979	0.885	0.090	>0.15	0.029	>0.25	0.231	>0.25
FP		0.893	0.335	0.189	>0.15	0.043	>0.25	0.316	>0.25

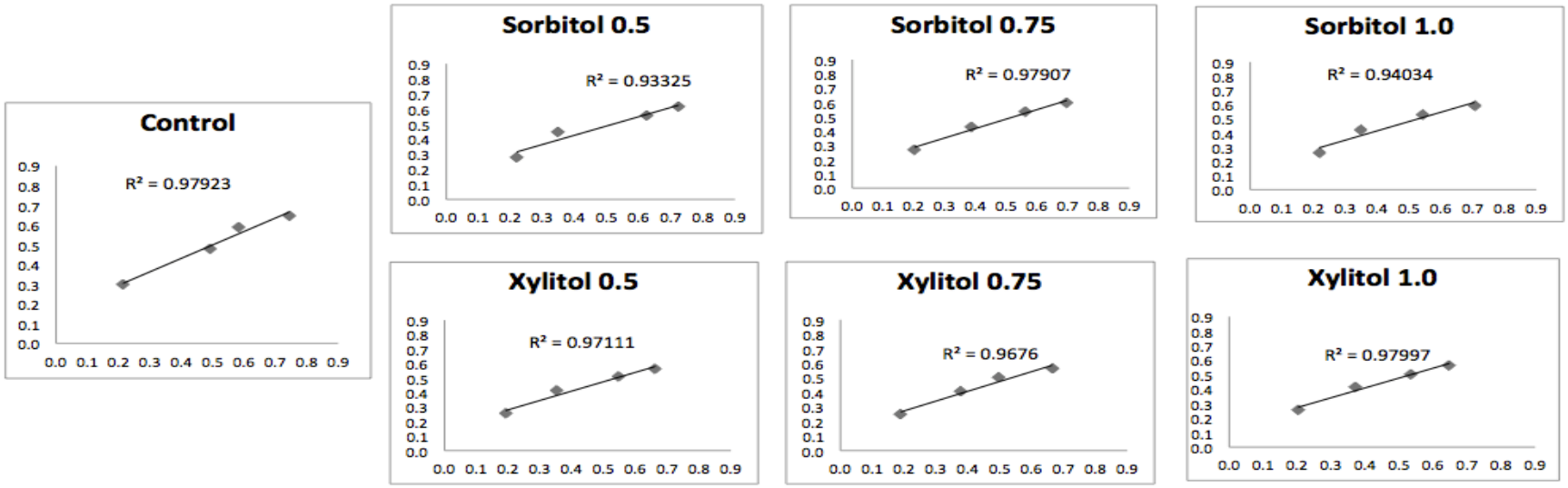
* Shapiro-Wilks *W*, Kolmogorov-Smirnov *D*, Cramer-von Mises W^2, and Anderson-Darling A^2. *P* values in bold show significant differences at α=0.05, indicating that the distribution of the residuals is non-normal.

APPENDIX 4.4: Mean values recorded in polysaccharide-based coatings

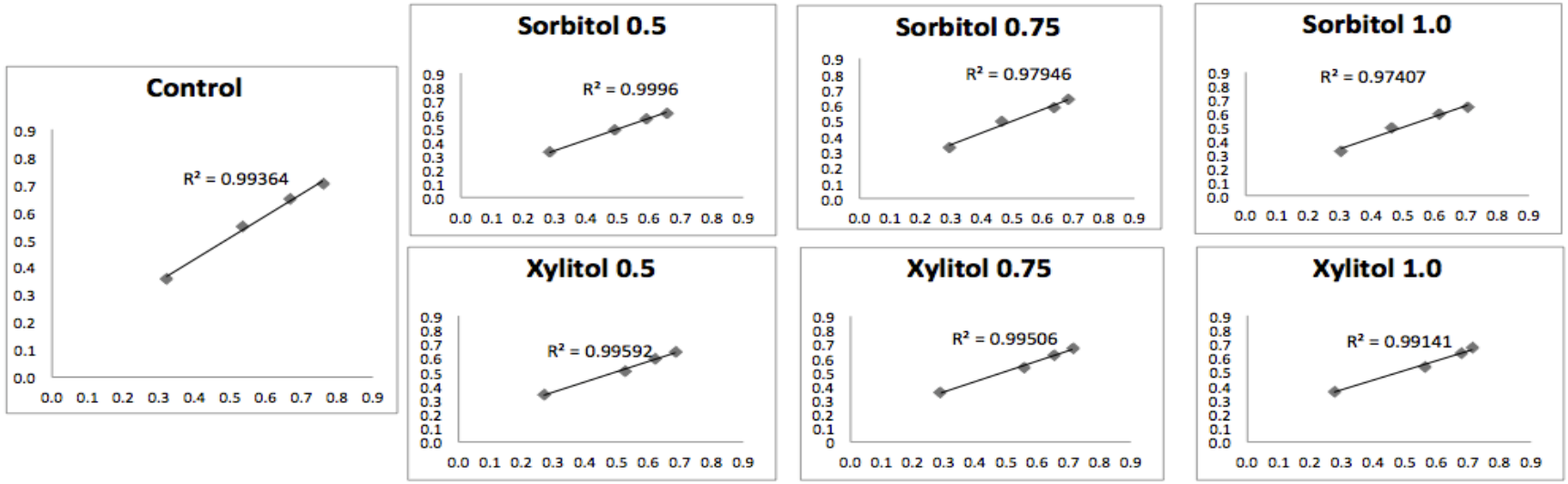
Parameter		Frying Time (s)	Coating Type					
			1% w/w Methylcellulose (MC)					
			Sorbitol (% w/w)			Xylitol (% w/w)		
			0.5	0.75	1.00	0.5	0.75	1.00
Moisture content	Final content (% w/w)	0	83.31	83.15	83.23	83.23	83.30	83.18
		60	55.15	53.76	52.98	56.27	54.36	55.72
		120	34.21	36.72	37.05	30.40	27.59	26.74
		180	23.98	19.71	21.52	20.86	18.10	15.32
		240	17.83	14.87	12.57	14.67	12.04	11.76
Film pick-up rate	(% w/w) per sample	60	3.68	2.32	2.85	3.68	2.70	0.96
		120	3.31	1.91	2.15	2.86	2.80	1.81
		180	3.36	1.72	3.00	2.97	2.66	1.86
		240	3.27	2.32	2.62	3.00	2.53	1.97
Fat	Final fat content (g w/w)	60	0.134	0.148	0.157	0.132	0.134	0.180
		120	0.187	0.194	0.202	0.226	0.226	0.242
		180	0.195	0.231	0.249	0.235	0.255	0.273
		240	0.211	0.242	0.272	0.259	0.301	0.307
	Fat reduction % (MC-NC)	60	38.82	32.62	28.45	39.73	38.97	18.01
		120	32.30	29.84	27.03	18.22	18.56	12.61
		180	34.14	22.12	15.90	20.74	14.09	7.81
		240	36.99	27.70	18.52	22.55	10.38	8.25
Color	L*	0	60.93	61.73	62.07	60.83	59.81	61.30
		60	60.37	59.63	60.38	60.64	59.97	59.38
		120	56.83	57.68	53.48	55.95	53.77	52.46
		180	48.24	53.35	48.43	46.78	46.67	47.81
		240	46.80	46.55	45.98	43.19	45.16	45.11
	a*	0	-2.77	-2.61	-2.49	-2.80	-2.34	-2.02
		60	-2.41	-2.32	-2.15	-2.35	-2.16	-1.99
		120	1.72	3.08	4.17	4.18	5.06	3.72
		180	6.36	5.60	7.65	5.92	5.33	5.24
		240	7.82	8.34	8.81	6.29	6.06	5.03
	b*	0	5.99	6.02	7.23	5.83	5.96	5.41
		60	6.23	6.86	8.86	6.11	6.14	6.76
		120	13.56	15.00	16.14	17.47	12.10	13.05
		180	13.29	15.72	13.28	10.10	11.42	12.06
		240	15.36	12.72	13.81	10.12	9.44	10.80
	ΔE	0	2.24	2.57	2.23	2.35	1.92	2.63
		60	2.92	3.15	2.12	4.93	3.13	1.89
		120	6.37	5.13	4.62	3.80	4.16	4.45
		180	5.77	7.75	3.56	8.31	4.81	5.59
		240	9.06	4.40	2.43	4.79	7.27	7.19
Texture	Load at maximum force (N)	60	0.48	0.43	0.41	0.44	0.61	0.57
		120	0.80	0.85	0.63	1.44	1.34	0.66
		180	1.75	1.61	1.95	1.93	3.00	1.85
		240	3.44	3.28	2.96	3.06	3.50	2.39
	Young's Modulus (N mm-1)	60	0.23	0.19	0.23	0.25	0.32	0.30
		120	0.24	0.51	0.23	0.63	0.54	0.35
		180	0.70	0.69	0.69	0.68	1.62	1.07
		240	1.18	1.15	1.37	1.28	3.00	2.68

Methylcellulose coating formulations mean values for all parameters studied in this study after 240 s frying time. Moisture loss, film pick-up rate, total fat up-take, color (L & ΔE), and texture development [Load at maximum force (N); Young's Modulus (λ).

APPENDIX 5.1: Data fit in proposed mathematical model for moisture loss responses in both WPI and MC coating formulations.

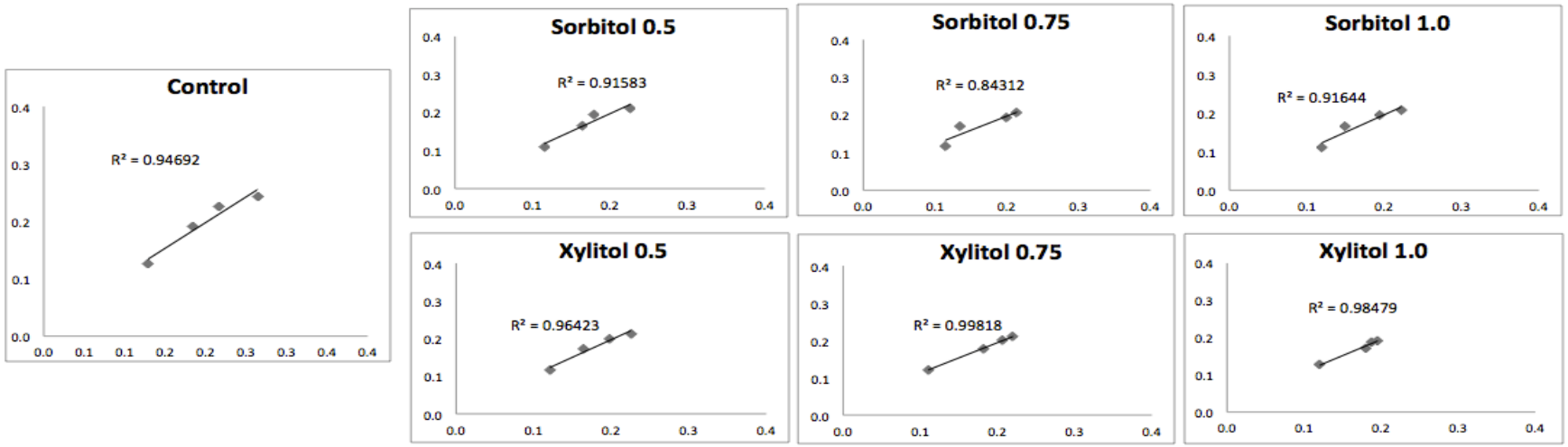


NC, WPI-0.5S, WPI-0.75S, WPI-1.0S, WPI-0.5X, WPI-0.75X, and WPI-1.0X.

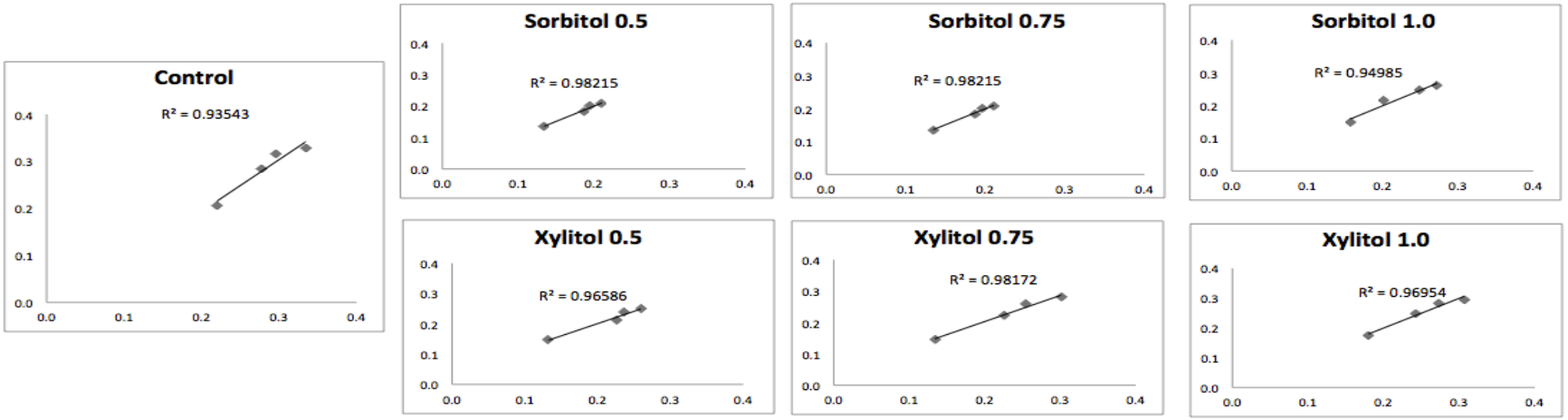


NC, MC-0.5S, MC-0.75S, MC-1.0S, MC-0.5X, MC-0.75X, and MC-1.0X.

APPENDIX 5.2: Data fit in proposed mathematical model for fat up-take responses in both WPI and MC coating formulations.



NC, WPI-0.5S, WPI-0.75S, WPI-1.0S, WPI-0.5X, WPI-0.75X, and WPI-1.0X.



NC, MC-0.5S, MC-0.75S, MC-1.0S, MC-0.5X, MC-0.75X, and MC-1.0X.