

EVALUATION OF THE MECHANISM OF OIL UPTAKE AND WATER LOSS DURING DEEP-FAT FRYING OF TORTILLA CHIPS

EVALUACIÓN DEL MECANISMO DE ABSORCIÓN DE ACEITE Y PÉRDIDA DE AGUA DURANTE EL FREIDO DE TORTILLA CHIPS

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Abstract

Simple equation $M_t/M_{\infty} = Kt^n$ is evaluated to describe the Fickian mechanism of oil absorption and water release during the frying of tortilla chips in nixtamalized samples with and without lime, where *n* is characteristic of the Fickian mechanism that changes with food physical and chemical properties. Moisture releases showed a Fickian mechanism form tortilla chips with lime (n = 0.5) due to water interaction with matrix components and non-Fickian for tortilla chips without lime (n = 0.6). Oil absorption on tortilla chips showed a Fickian mechanism for nixtamalization with lime n = 0.4 and for nixtamalization without lime, one of n = 0.3. The larger water diffusion coefficient (D_w) in the tortilla chip compared to that of oil diffusion (D_o) indicates that water release and oil absorption during frying is not a simultaneous event. During tortilla chip frying, water release fallowed a chemical mechanism due to the interactions of matrix component materials with n > 0.5, while oil absorption presented physical mechanism n < 0.5 that was time independent. Diffusion exponent *n*, was related to the water released or oil absorption mechanisms.

Keywords: Fickian mechanism, diffusion coefficient, water release, oil absorption, tortilla chips.

Resumen

Evaluación de una ecuación simple $M_t/M_{\infty} = Kt^n$ para describir el mecanismo Fickiano o no Fickiano de absorción de aceite y liberación de agua durante freído de tortillas chips nixtamalizada con y sin cal, M_t/M_{∞} es la parte fraccional de la absorción o liberación del soluto, n es el exponente de difusión característico del mecanismo Fickiano que cambia con la parte física y química del alimento. Para un mecanismo Fickiano, el valor del exponente es $n \le 0.5$, para uno no Fickiano 0.5 < n < 1 en el caso de una hoja plana. La liberación de agua para una tortilla chip nixtamalizada con cal presentó un mecanismo Fickiano (n = 0.5) atribuido a interacciones del agua con los componentes de la matriz y mecanismo no-Fickiano para tortilla chip sin cal (n = 0.6). La absorción de aceite en tortilla chip mostró un mecanismo Fickiano en la nixtamalización con cal (n = 0.4) así como para el cocimiento sin cal (n = 0.3). Un coeficiente de difusión del agua mayor, comparado con el del aceite indica que son eventos no simultáneos durante el freído. Durante el freído de la tortilla chip, la liberación de agua siguió un mecanismo químico debido a las interacciones con los componentes de la matriz del material con n > 0.5, mientras, la absorción de aceite presentó un mecanismo físico n < 0.5, que fue independiente del tiempo. El exponente de difusión, n se relacionó con los mecanismos de liberación de agua y absorción de aceite.

Palabras clave: mecanismo Fickiano, coeficiente de difusión, liberación de agua, absorción de aceite, nixtamalización, tortilla chips.

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1 Introduction

Deep-fat frying is one of the most important unitary operations in food processing. Frying is a traditional cooking method to achieve desirable sensory attributes such as flavor and texture in variety of foods (Blumenthal and Stier, 1991; Mohan et al., 1995). During frying, heat affects the moisture and oil transfer in a quasi-simultaneous event in which moisture leaves the food in the form of vapor, while the oil is absorbed (Fan et al., 2005; Ding-Bing et al., 2013). Mass transfer is characterized by starch, soluble materials, and water escaping from the products, while oil is absorbed into the food (Kochhar and Gertz, 2004). In deep frying, this leads to the formation of a crispy hard crust and, due to the high frying temperatures that promote moisture loss, chemical changes such starch gelatinization, protein denaturation, caramelization, and the Maillard reaction, which turns the crust to a golden-brown color and provides it with desirable flavors (Kochhar and Gertz, 2004; Goyary et al., 2015). There are several factors that affect heat and mass transfer, among these the thermal and physicochemical properties of the food, and the oil type, the geometry of the food, and the temperature of oil are determinants in the final quality (Krokida et al., 2000). Several authors indicated that the amount of oil absorbed was related to particle size, initial moisture content (Moreira et al., 1997), and pores size distribution (Moreira et al., 1997; Bouchon et al., 2003). As well as the oil temperature (Moreira and Barrufet, 1996) in tortilla and potato chips. As people become more health conscious and pursue healthier food, the trend of consumers has changed to low-fat fried food over the past years (Stier, 2004; Bouchon and Pyle, 2004). A better understanding of the relationship to different parameters of the deep-fat frying should provide ways to optimize the frying process, thus controlling oil pickup (Ni and Datta, 1999). To overcome that problem, several papers have been written, mainly using Fick?s law of diffusion for modelling and predicting the moisture loss and oil absorption during the deep-fat frying of potato and tortilla chips (Tomasula and Craig, 1991; Chen and Moreira, 1997). The majority of the models that have been developed are based on solutions of the Fickian diffusion equation published elsewhere (Crank, 1975; Ritger and Peppas, 1987a). Diffusion can be described using Ficks second law, and there are various ways to apply the respective equation.

First, it is important to consider geometry. It may be assumed that dimension transport on a thin slab will result in a rather exponential mathematical series, but this approach is only valid for flat, planar samples. In addition, it is important to decide whether to assume constant or non-constant diffusivity. Other researchers had been applying a similar Fick's law mechanism to understand the release of drugs from several medicament matrices. Some of these pioneering works were done by Higuchi (1961; 1963), who studied the release rate of medicament drugs in suspension, and proposed an equation that shows the instantaneous rate of absorption at time t and that it can regulate the release of drugs from such a preparation by controlling different variables. This allowed the quantification of drug release from thin film. This equation was extended to other geometrics, and related theories have been proposed (Siepmann and Peppas, 2011). Based on the studies of Higuchi (1961; 1963) and Crank, (1975), other authors (Ritger and Peppas, 1987a) introduced a simple and convenient Power law relationship, $M_t/M_{\infty} = Kt^n$, which may be used to describe the Fickian and non-Fickian release behavior of swelling-controlled release systems. For the Fickian mechanism, the exponent (n) is ≤ 0.50 , and non-Fickian is 0.5 < n < 1 for slabs. The diffusional exponent, (n) simplifies the analysis of the frying process due to an important indicator of the mechanism of transport of a material through the sample. Tortilla chips are the second place of preferred snacks, especially by children, only behind potato products. The high health risks involved in the consumption of high levels of fat in those products, require a better understanding of the oil absorption and water release mechanisms in order for reducing the levels of oil in this compound specially in tortilla chip that are consumed by a large segment of the population (Topete-Betancourt et al., 2019). The main objective of this work was to evaluate the Fickian mechanism of oil absorption and water loss during the deep-fat frying as affected by the nixtamalization process, through the proposal of a simple Power law equation, which can be used to analyze and compare the simultaneously released water and oil absorption.

2 Materials and methods

2.1 Tortilla chips preparation

The tortilla chips were prepared with the traditional nixtamalization process with lime (TD) (1% w/w) food grade (Calcium hydroxide Ca(OH)₂, purchased in Alquimia Mexicana, Mexico, D.F.) and without lime using only water (TC). White maize (1 Kg, Purchased in the commercial market in Queretaro, Mexico.) was added to a Ca(OH)₂ solution in a proportion of 2:1 w/v, then mixture was cooked for 25 min at 95 °C, removed from heat, and steeped for 16 h at room temperature. The cooked kernels with 44 % moisture content for lime and 40 % moisture without lime (nixtamal) were ground in a stone mill to obtain fresh masa with a 607 μ m of particle size recommended for the tortilla chip. The masa was mixed for 3 min with distilled water at room temperature (100 ml/500 g masa) until achieving the adequate consistency. The masa nixtamalized with lime showed a 46 % moisture and 47 % moisture for masa without lime. Then, the masa from both treatments was shaped into the form of flat discs using a small-roll tortilla machine (Casa González, Mexico City, Mexico) with a gap of 0.172 mm and a weight of 14 g. The tortilla was baked on an IR oven (Black & Decker) at 260°C for 45 s (15 s, 15 s, 15 s, by each side), followed by 15 min resting time at room temperature. It was used a triangular (5x5x5 cm) mold to shape the baked tortilla into tortilla chip with 1.65 g weight average and 0.180 mm of thickness. For each experimental time were used batches of 100 g (60 samples) of tortilla chip to fry in 3 L corn oil (Mazola) at 185°C using a fryer of 5 L capacity (T-fal Filtra Pro Inox) for 0, 5, 10, 15, 30, and 60 s to determine the amount of water loss and oil absorption in the chips as a function of frying time.

2.2 Moisture content

The moisture content of the tortilla chip was determined by the approved 44-19.01 method (AACC International, 2010). From batch of tortilla chip elaborated with different nixtamalization process with lime and without no lime, ten pieces (18 g) were taken randomly after reaching the room temperature at 25°C and kept inside a polyethylene bag. The tortilla chip pieces were grinded using a coffee grinder (GX4100) and by triplicate 2 g samples were placed in a force air oven at 105°C for 24 h.

2.3 Oil content

Total content oil in the tortilla chip was determined by the approved 30-10.01 method (AACC International, 2010) with acid hydrolysis. Briefly, from batch of tortilla chip elaborated with treatment above mentioned, ten pieces (18 g) were taken randomly after reaching the room temperature at 25°C and kept inside a polyethylene bag. The tortilla chip pieces were grinded using a coffee grinder (Model-GX4100). Place 2 g grinded tortilla chip in 500 ml beaker, add 10 ml HCl 6N solution, and stir during 1 min to moisten all particles. Add 2 ml ethyl alcohol 96%, mix well for 1 min. set beaker in water bath held at 70-80°C, for 30 min, shake it intervals 10 min. After this time, add 10 ml ethyl alcohol 96% shake and cool at room temperature. Transfer mixture to separation funnel. Add 25 ml ether in three portions, shake vigorously 1 min and let repose 3 min after each portion. Add 25 ml petroleum ether in three portions, shake vigorously 1 min and let repose 3 min, after each portion. Let stand 20 min until phase upper liquid is practically clear. Draw off clear ether solution through filter papers 42 (125 mm diameter) into previously weighed 500 ml round-bottom flask. Evaporate ether slowly on steam bath into laboratory chamber. Dry fat in drying oven at 100 °C, for 100 min. Remove the round-bottom flask and led stand in air to constant weight.

Calculation

$$\%Fat = \frac{\text{Weight of fat (corrected for blank}}{\text{Weight of sample}} \times 100$$

2.4 Models to describe water evaporation and oil absorption on a plane sheet

The majority of the models that have been developed are based on solutions of the Fickian diffusion equation published elsewhere (Crank, 1975; Ritger and Peppas, 1987a; 1987b). Fickian diffusion of water evaporation and oil absorption in thin-slab was considered in one dimension, and the system is initially maintained at a constant uniform oil concentration C_1 and its surfaces are kept at a constant oil concentration, C_0 . This situation corresponds to typical experimental conditions for sorption and desorption experiments and is referred to as the perfect sink condition. For assumed constant oil diffusion in the *x* direction, Fick?s second law, along with the appropriate initial and boundary condition, may be written as:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \tag{1}$$

Where

$$t = 0 - l/2 < x < l/2 \quad C = C_0$$

$$t > 0 \quad x = \pm l/2 \quad C = C_1$$

The solution to Fick's law in the form of a trigonometric series under the previously specified conditions is given by Crank (1975):

$$\frac{M_t}{M_{\infty}} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 \pi^2} \exp\left(\frac{-D(2n+1)^2 \pi^2}{4l^2} t\right)$$
(2)

Where M_t is defined as the mass of the tortilla chip at time t, and M_{∞} is the mass of tortilla chips at infinity time or when the curve becomes asymptotic. D is the diffusion coefficient. An alternate solution to Eq. (1) that is useful for the interpretation of short-time, as presented in the few seconds of the deep frying, is that behavior is given in the form of an error function series:

$$\frac{M_t}{M_{\infty}} = 4 \left[\frac{Dt}{l^2} \right]^{1/2} \left[\frac{1}{\pi^2} + 2 \sum_{n=1}^{\infty} (-1)^n ierfc \frac{nl}{2\sqrt{Dt}} \right]$$
(3)

Where ierfc represents the integrated complementary error function of *x*. For "small times", i.e., small values of the dimensionless time *t*, defined as $4Dt/l^2$, Eq. (3) can be approximated by Ritger and Peppas (1987) as follows:

$$\frac{M_t}{M_{\infty}} = 4 \left[\frac{Dt}{\pi l^2} \right]^{1/2} \tag{4}$$

As indicated by Eq. (4), the Fickian diffusional coefficient in the absorption of oil and the desorption of water from a thin slab is characterized by an initial \sqrt{t} time dependence of the released of water and the absorption of oil. The generalized expression of the previous Power-law equation can be written as:

$$\frac{M_t}{M_{\infty}} = Kt^n \quad or \quad \frac{M_t}{M_{\infty}} = K_1 + Kt^n \tag{5}$$

Where *K* is a constant incorporating characteristic of the macromolecular network system and K_1 is the initial value when $M_t/M_{\infty} \neq 0$ Ritger and Peppas (1987a) for the case of the release of the water, and n is the diffusional exponent, which is indicative of the Fickian mechanism. The Fickian diffusion mechanism is defined by $n \leq 0.50$, and being within the range of 0.5 < n < 1.0 indicates a non-Fickian mechanism. The semi-empirical Power law Eq. (5) can be used to relate the amount of released water and oil absorption as an exponential function of the release and absorption time. Characteristic diffusional exponents for Fickian diffusional release have been defined in each case, for the fitting of the first 60% of the release curve for long times (Ritger and Peppas, 1987b). However, in the case of tortilla chips, the processing time during deep frying is relatively short; thus, the majority of the curve is valid for 100% of the event. Also, we assumed that the material had a plane sheet shape with a thickness of *l*. We proposed that the equivalent thickness of the plane sheet is the mean value of the material slides. For this experiment, the equivalent thickness of the tortilla chip was 1.32 mm. Thus, to compute the coefficient diffusion, it was estimated with this value.

2.5 Statistical analyses

From experimental release, water and oil absorption data, together with Eq. (2), the Diffusion coefficient of water release (D_w) and the Diffusion coefficient of oil absorption (D_o) were calculated using Mathematica ver. 10.4 software (Wolfram Research Company).

From fractional release, water and oil absorption data were exponent diffusion calculated using a nonlinear regression performed with Prism ver. 5 for Windows (PrismPad Software, Inc., La Jolla, CA, USA). Two replicates were used for each treatment.

3 Results and discussion

3.1 Water loss during deep fat frying of tortilla chips

Fig. 1 shows differences between the initial moisture in the traditional tortilla chip (TD) (Fig. 1A) and in the tortilla chip control processed only with water (TC), with higher moisture for TD (Fig. 1A). A possible explanation of this difference is because traditional nixtamalization represents an alkaline process that induces pericarp hydrolysis, starch gelatinization, protein solubilization, and lipid saponification which facilitates water absorption (Santiago-Ramos et al., 2018), Thus contrary to what occurs in corn processed without lime $[Ca(OH)_2]$ in which the pericarp exhibits less damage, due to the neutral condition, where the lipid saponification and protein solubilization are probably less. However, both treatments demonstrated the typical diffusion curve of Fick's law, although with significant differences among them.



Fig. 1. Moisture loss of tortilla chip during deep frying at temperature of 185°C. A). Tortilla chip nixtamalized TD with 3% lime w/w. B). Tortilla chip nixtamalized TC without lime (Control).

Moisture loss from TC behavior was faster than that of TD, due to that TC maintains a short curve shape from zero to 30 s, and after this point, it becomes straight, meaning that the water loss reached equilibrium in a shorter time compared with TD that, after 30 s, exhibits an asymptotic tendency, requiring more time to reach equilibrium during the frying. It is noteworthy that the TD had a greater loss of water from zero to 15 s than the TC, but oil absorption does not follow the same behavior. These data are in agreement with those reported by Moreira et al. (1995), among others, which indicated that water release was faster during the initial seconds due to water vapor expansion and an increase of porosity during the frying. However, the interaction of the water with other macromolecules has not been discussed during the deep-fat frying.

3.2 The mechanism of water release during deep frying of tortilla chip

Fig. 2 depicts the moisture loss data presented as a percentage and M_t/M_{∞} concentration ratio. The data exhibit the same trend using the experimental time defined by Ritger and Peppas, (1987a) as $t^{1/2}$ similar tendency to $(Dt)^{1/2}/l$ where $D = D_w$ is the water diffusion coefficient, which it was calculated with the experimental data applying Eq. (2), and, l

was the average thickness of the tortilla chip (1.32 mm). The Fickian diffusion coefficient for tortilla chip nixtamalized with lime was (TD) of $D_w = 8.40 \times 10^{-6}$ m²/s, and for tortilla chip cooking without lime (TC), was $D_w = 8.56 \times 10^{-6}$ m²/s. Both treatments appear to have the same diffusion coefficient values with the plotting tendencies of Fig 2. Higher values of water diffusion coefficients were reported by several authors (Moreno-Castro et al., 2015; Moreira et al., 1995; Ritger and Peppas, 1987a; Tong and Lund, 1990; Ni and Datta, 1999; Mandema and Zeldenrust, 1977; Halder et al., 2007; Enscore et la., 1977), probably due to the different materials and different particle size employed. Fig. 2 presents the moisture loss behavior using the coefficient diffusion, plotting for the TD (Fig. 2A) and TC (Fig. 2B), as well as using the square root of the frying time for TD (Fig. 2C) and TC (Fig. 2D). Each treatment demonstrated a similar tendency, either using the coefficient diffusion values or the square root of the frying time, which support a simple way to analyze moisture loss applying the square root of the frying time instead of the coefficient diffusion calculated from a complicated equation (Eq. 2). On the other hand, to determine the diffusion mechanism in both treatments (with and without lime), this considered constant coefficient diffusion, thin slab geometry, and fractional concentration.

Table 1. Relation between release exponent (n) values for the Ritger and Peppas Eq. (5) and type of mechanism depending on the geometric shapes.

Release exponent (<i>n</i>)			Release mechanism
Thin film	Cylindrical	Sphere	
0.5	0.45	0.43	Fickian diffusion
0.5< n <1.0	0.45< n <0.89	0.43< n <0.85	Non-Fickian diffusion
1.0	0.89	0.85	Case II transport



Fig. 2. Loss of water during deep frying at 185° C of tortilla chip. Continuous line is the experimental data. Dotted black line is the fitted data to the Eq. (5). Dotted gray lines are the 95% confidence band. A) Tortilla chip with 3% lime and water Fickian diffu sion coefficient D_w of $8.40 \times 10^{-6} \text{ m}^2/\text{s}$. B) Tortilla chip without lime and water Fickian diffusion coefficient D_w of $8.56 \times 10^{-6} \text{ m}^2/\text{s}$. C) Tortilla chip with 3% lime and D) Tortilla chip without lime using square time. The thickness of sample was 1.32 mm.

The Peppas model based on the Power law Eq. (5) was used to estimate the diffusional exponent (n) indicative of the type of transport mechanism for different geometrical shapes as shown in Table 1.

The values obtained for TD were n = 0.5 ($R^2 = 0.98$) and n = 0.6 ($R^2 = 0.97$) for TC. The TD possessed a Fickian diffusion and TC a non-Fickian diffusion transport, and those values were in agreement with the water loss behavior and that TC reached the equilibrium faster than TD. Although,

TC did not meet with Fick second diffusion law due to straight behavior, the data were within the 95% confidence band of the regression curves. Fig 3. Illustrates how the water molecules are surrounded by macromolecules, forming multilayers, as well as interacting with the internal macromolecules through the hydrophilic groups and covering the vacant spaces between the chains with a configuration of crosslinking. During the first seconds of frying, the external multilayer of water is released during the vapor phase



Fig. 3. Tortilla chip. Water release from water particle layer during first seconds and water loss from inside the particles.

from the tortilla chip network, which is considered to be the same in both treatments, with a similar rate of moisture reduction ranging from 20% - 10%.

Regarding the TD, the water release behavior showed less variation before reaching the moisture equilibrium, specifically at 7% to 2%. Our hypothesis indicates that during frying time, the water molecules made a strong attraction force through the hydrophilic groups (-NH₂, = NH, -OH, and -COOH) present in the macromolecules, such as protein denaturalization, arabinoxylans generated by the hydrolysis of the corn pericarp in the masa, and mostly, starch gelatinization, which lead the formation of different water layers in these groups. All of these macromolecules form a continuous phase and a strong network that reduce the water release in TD compared with that of TC during frying. This hypothesis could be supported by Soppirnath and Aminabhavi, (2002), who mentioned that water uptake in hydrogels depends on the degree of hydrodynamic free volume and the availability of hydrophilic functional groups for the water to establish hydrogen bonds, and their transport depends on the degree of cross-linking. Maitra and Shukla, (2014) reported that polymer chains in the presences of water might cross-linked to form a hydrogel, and that the water occupies the voids in the network. Korsmeyer

and Peppas, (1981) concluded that the cross-linked macromolecular structure affected the solute diffusion process. Chou and Morr, (1976) suggested that the interaction of water with protein molecules exhibits different properties because this is directly related to the manner in which the protein interacts with the water. This is described as structural monolayers and the factors that affect the protein-water interactions involve distinct conditions such as pH, salt, and temperature. Traditional nixtamalization has all of the factors mentioned previously (Arámbula et al., 2000). An increase in the formation of water multilayers around the macromolecules, using lime might be expected compared with cooking corn with water (TC). This is because it is known that lime promotes changes in the internal matrix, such as cross-linking between amylose/amylopectin with Ca2+ (Cornejo-Villegas et al., 2018; Bryant and Hamaker, 1997). Grabowska et al. (2015) found that the introduction of Ca²⁺ ions caused the formation of cross-linking ionic bonds within carboxyl and carboxylate groups. Moussa and Cartilier (1996) reported the use of amylose to obtain different degrees of cross-lining in alkaline medium in order to observe the swelling and solvent concentration gradients into these matrices. Furthermore, the calcium is the most widely employed

cation to obtain the formation of cross-linking in the gel network, which decreases permeability (Aslani and Kennedy, 1996). Table 2 presents the values for the diffusion exponent (n) and the diffusion coefficient (D) for water release during tortilla chip frying, and these values were in agreement with those reported by Soppirnath and Aminabhavi, (2002), who suggested that the D_w and n values decreased when the crosslinking of the network polymer increased. Gallegos-Marin et al., (2016) mentioned that water diffusion coefficients are related to the increase or decrease of pores in the matrix material due to changes in the starch granule. It has been very often pointed out that the properties of water molecules in polymer gels are different from those of free water, and that the degree of the differences is dependent upon the degree of cross-linking.

3.3 Effect of the different nixtamalization conditions on the diffusion coefficients

Fractional data deriving from water release were plotted using the D_w , as shown in Figs. 2A and 2B. both treatments appear to have the same behavior from $M_t/M_{\infty} = 0.4$ to 0.15 but, during the last three points, the treatments exhibited an inflection. TD changed to a slightly curve compared with TC, that showed a straight reached the equilibrium before that TD. The diffusion coefficients calculated for TC was $D_w = 8.56 \times 10^{-6} \text{ m}^2/\text{s}$, while for TD, this was $D_w = 8.40 \times 10^{-6} \text{ m}^2/\text{s}$. These values might explain the similarity on behavior; nevertheless, they were some relatively different values than those reported in the literature. A possible explanation could be due to different conditions, such as higher particle size in masa, $185^{\circ}C\pm3$ oil temperature, and the 1.32 mm slabs thickness in the tortilla chip. This might supported by Chen *et al.* (2009), who measured the D_w from yellow-dent corn kernel components at different temperatures, including 35°C, 45°C, 55°C, and 65°C, for each part, and reported, for all temperatures used: $D_w = 8.79 \times 10^{-6} \text{ m}^2/\text{s}$ in the germ, $D_w =$

 3.84×10^{-6} m²/s to 3.73×10^{-6} m²/s in the pericarp, and $D_w = 4.08 \times 10^{-6} \text{ m}^2/\text{s}$ in the hard endosperm, respectively. Flores-Martinez et al., (2018) mentioned that diffusion coefficient of essential oil increases when the temperature is heightening and reported a $D = 5.3586 \times 10^{-11} \text{ m}^2/\text{s}$ at 5°C, while at 25°C was D_w $= 5.2578 \times 10^{-8}$ m²/s. Moreira *et al.* (1995) measured (D_w) at different oil temperatures in tortilla chips elaborated with nixtamalized corn flour and reported a $D_w = 9.348 \times 10^{-8} \text{ m}^2/\text{s}$ for oil heated at 190°C and a $D_w = 5.408 \times 10^{-8} \text{ m}^2/\text{s}$ for oil at 150°C. On the other hand, Olguin-Rojas et al., (2019) mentioned that the differences between diffusion coefficient values possibly is due to sample shape and size. Contreras-Jiménez et al. (2014) calculated the D_w for corn grits with and without lime at room temperature and reported values of $D_w = 2.24 \times 10^{-10} \text{ m}^2/\text{s}$ without lime, and $D_w=2. \times 10^{-10} \text{ m}^2/\text{s}$ with lime. Arámbula et al. (1999) reported the D_w in masa prepared with extruded corn flour gums additives were incorporated before and after the extrusion process; the D_w was 4.35×10^{-9} m²/s and 4.65×10^{-9} m²/s respectively, and mentioned that this difference was due to the water retention promoted by the additions of gums, which could have engendered more interaction with other molecules such as: proteins, lipids and carbohydrates.

3.4 Oil absorption during deep fat frying

Regarding the oil absorption, Fig. 4 shows the percentage of oil absorption of the samples with lime and control vs. time. The nixtamalized sample (TD) showed a characteristic Fickian curve compared with tortilla chip prepared without nixtamalization (TC, control). The sample absorbed about 18% of oil during the first 5 s of frying compared to the sample nixtamalized with lime that required 20 s to absorb the same amount of oil. However, the oil absorption during frying increases as the initial moisture of the tortilla chip decreases. In this work, the initial moisture of the TD was higher than that of the TC, although the final tortilla chip oil at the same frying time was nearly the same in both samples. However, TD appears to have the capacity to absorb more oil.



Fig. 4. Oil absorption of tortilla chip during deep frying at 185°C. A). Tortilla chip nixtamalized TD with 3% lime w/w. B). Tortilla chip nixtamalized without lime TC (Control).

3.5 Mechanism of oil absorption during deep fat frying

Fig. 5B shows that, during the first 5 s, the absorption of oil in TC is faster than the traditional nixtamalized sample (TD) (Fig. 5A). Although the mechanism of oil absorption in tortilla chips comprises an important quality parameter, there is a little, information reported about the oil diffusion coefficient (Do). This may be due to the difficulty in performing oil extraction compared with the water diffusivity coefficient D_w . The oil diffusion coefficients (Do) calculated from Fick's law were as follows: TD had DO: 4.05×10^{-7} m^2/s , and TC had Do: $7.71 \times 10^{-7} m^2/s$. This means that the oil absorption is a faster event in TC than of oil absorption in TD, and in the last period of frying TC presents saturated absorption. However, TD demonstrated slower oil absorption. This tendency was similar to that of the data reported by Chen and Moreira, (1997) and Krokida et al. (2000). Moisture loss decreases as the oil absorption increases; thus, there is a relationship between loss water and oil absorption in the same sample. Several authors indicated that, during frying, heat affects moisture and oil transfer, in which moisture leaves the food in form of vapor, while the oil is absorbed (Fan et al., 2005; Ding-Bing et al., 2013). The larger water diffusion coefficient $D_w = 8.56 \times 10^{-6} \text{ m}^2/\text{s}$ in the tortilla chip compared with the oil diffusion coefficient Do = 7.71×10^{-7} m²/s indicates that water release and oil absorption during deep frying is not a simultaneous event (Rubnov and Saguy, 1997). Calculating diffusion coefficients from the equations and data reported by Chen and Moreira, (1997), showed similar tendencies as the present work with differences of D_w of 1.8×10^{-8} m^2/s vs. Do of $3.02 \times 10^{-12} m^2/s$ for the tortilla chip, where the D_w was higher than the Do.

Fig. 5A and 5B show the experimental data that fitted the (Ritger and Peppas, 1987a) for obtaining the value of diffusional exponent (n), where the oil absorption in TC is faster than in TD. Similar trends were obtained with Eq. (5) using frying time (Fig. 5C and D), where the exponents were n=0.4 and n=0.3 for TC and TD, respectively. These results were in agreement with those reported in the literature in relation to water content and the oil absorption, and these might be interpreted such as the amount of easily lost water and the oil absorption suggested by Krokida et al. (2000). However, there was an inverse tendency of diffusion exponents (n) compared to the water exponents shown in Table 2. This fact indicates that the interaction mechanism with hydrophilic groups is only valid for water because the hydrophobic character oil makes the oil absorption relaying mainly on the void spaces left by the evaporation of the water during frying.

Conclusions

The M_t/M_{∞} fractional data as a function of time square can be a practical way to analyze Fick's diffusion mechanism in food systems such as tortilla chips. Water release was faster in the tortilla chip without lime than in that nixtamalized with lime, although nixtamalization with lime followed a Fickian mechanism and non-lime samples fitted a non-Fickian diffusion. Oil absorption in nixtamalization with lime was slower than nixtamalization without lime, this showed Fickian diffusion.



Fig. 5. Oil absorption during deep frying at 185°C of tortilla chip. Continuous line is the experimental data. Dotted black line is the fitted data to the Eq. (5). Dotted gray line are the 95% confidence band. A) Tortilla chip TD with 3% lime and with Fickian dif fusion coefficient D_o of 4.60 x10⁻⁷ m²/s. B) Tortilla chip TC without lime and with Fickian diffusion coefficient D_o of 7.71 x10⁻⁷ m²/s. C) Tortilla chip with 3 % lime using square time and D) Tortilla chip without lime using square time. The thickness of both samples was 1.32 mm.

According to the results, the mechanisms of water release and oil absorption during frying are not simultaneous events, due to, that the water release diffusion coefficient was higher than the oil absorption diffusion coefficient, while, the diffusional exponent for the water release was a non-Fickian mechanism and for the oil absorption was a Fickian mechanism. This model can be used to analyse the different frying parameters, such as the nixtamalization process. The behavior of the water and that of oil diffusion continues to be an open topic that, in this study, was investigated by means of the transport equation. Because people are becoming more health conscious, oil absorption in the tortilla chips is an important quality parameter; however, to our knowledge, there is little, or a lack of, information about the oil absorption diffusion coefficient and the mechanism involved in it.

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Nomenclature

- M_t the mass of tortilla chip at time t (s)
- M_{∞} the masa of tortilla chip at infinity (s)
- 1 thickness of the sample (mm)
- D diffusion coefficient (m^2/s)
- D_w diffusion coefficient of water release (m²/s)
- Do diffusion coefficient of oil (m^2/s)
- t time (s)
- Co constant oil concentration
- C1 uniform oil concentration

- K constant characteristic of the macromolecule network
- n diffusion exponent

References

- AACC International. (2010). *Approved Methods of Analysis*, 11th Ed. Methods 44-19.01, 30-10.01. Available online only. AACC Int: St. Paul, MN, USA.
- Arámbula, V. G., Mauricio, S. R. A., Figueroa, J. D. C., González-Hernández, J., and Ordorica, F.C.A. (1999). Corn masa and tortilla from extruded instant corn flour containing hydrocolloids and lime. *Journal of Food Science* 64, 120-124.
- Arámbula, V. G., Yáñez-Limón, J. M., Vorobiev-Vasilievich, Y., and González-Hernández, J. (2000). Effective moisture diffusion coefficient of nixtamalized corn masa by extrusion. *Agrociencia 34*, 717-727
- Aslani, P. and Kennedy, R. A. (1996). Studies on diffusion in alginate gels. I. Effect of crosslinking with calcium or zinc ions on diffusion of acetaminophen. *Journal of Controlled Release* 42, 75-82.
- Blumenthal, M. M. and Stier, R. F. (1991). Optimization of deep-fat frying operations. *Trends in Food Science and Technology* 2, 144-148.
- Bouchon, P., Aguilera, J. M. and Pyle, D. L. (2003). Structure oil absorption relationships during deep-fat frying. *Journal of Food Science* 68, 2711-2716.
- Bouchon, P. and Pyle, D. L. (2004). Studying oil absorption in restructured potato chips. *Journal of Food Science* 69, 115-122.
- Bryant, C. M. and Hamakar, B. R. (1997). Effect of lime on gelatinization of corn flour and starch. *Cereal Chemistry* 74, 171-175.
- Chen, Y. and Moreira, R. G. (1997). Modelling of a batch deep-fat frying process for tortilla chips. *Food and Bioproducts Processing* 75, 181-190.

- Chen, G., Maier, D. E., Campanella, O. H. and Takhar, P. S. (2009). Modeling of moisture diffusivities for components of yellow-dent corn kernels. *Journal of Cereal Science* 50, 82-90.
- Chou, D. H. and Morr, C. V. (1976). Protein-water interactions and functional properties. *Journal* of the American Oil Chemists' Society 56, 53-62.
- Contreras-Jiménez, B., Gaytán-Martínez, M., Figueroa-Cárdenas, J. D., Avalos-Zúñiga, R. A. and Morales-Sánchez, M. (2014). Effect of steeping time and calcium hydroxide concentration on the water absorption and pasting profile of corn grits. *Journal of Food Engineering 122*, 72-77.
- Cornejo-Villegas, M. A., Rincon-Londoño, N. R., Real-Lopez, A. and Rodríguez-Garcia, M. E. (2018). The effect of Ca²⁺ ions on the pasting, morphological, structural, vibrational, and mechanical properties of corn starchewater system. *Journal of Cereal Science* 79, 174-182.
- Crank, J. (1975). *The Mathematics of Diffusion*. 2nd Ed., Clarendon Press.
- Ding-Bing, H., Fei, X., Tse-Chao, H. and Xiao-Yan, S. (2013). Oil absorption mechanism of fried food during cooling process. *Journal of Food Process Engineering* 36, 412-417.
- Enscore, D. J., Hopfenberg, H. B. and Stannett, V. T. (1977). Effect of particle size on the mechanism, controlling n-hexane sorption in glassy polystyrene microspheres. *Polymer 18*, 793-800.
- Fan, L. P., Zhang, M., Xiao, G. N., Sun, J. C. and Tao, Q. (2005). The optimization of vacuum frying to dehydrated carrot chips. *International Journal* of Food Science and Technology 40, 911-919.
- Flores-Martinez, N. L., Perez-Perez, M. C. I., Oliveros-Muñoz, J. M., Lopez-Gonzalez, M. L. and Jimenez-Islas, H. (2018). Estimation of diffusion coefficients of essential oil of *Pimenta dioica* in edible films formulated with *Aloe vera* and gelatin, using Levenberg-Marquardt method. *Revista Mexicana de Ingenieria Quimica 17*, 485-506.
- Gallegos-Marin, I., Mendez-Lagunas, L. L., Rodriguez-Ramirez, J. and Martinez-Sanchez,

www.rmiq.org

C. E. (2016). Structural properties changes during osmotic drying of plantain (*Musa paradisiaca* AAB) and its role on mass transfer. *Revista Mexicana de Ingeniería Química 15*, 441-456.

- Goyary, J., Kumar, A. and Nayak, P. K. (2015). Changes of quality parameters of soybean oil in deep fat frying: A review. *Journal of Basic and Applied Engineering Research* 2, 951-954.
- Grabowska, B., Sitarz, M., Olejnik, E. and Kaczmarska, K. (2015). FT-IR and FT-Raman studies of cross-linking processes with Ca²⁺ ions, glutaraldehyde and microwave radiation for polymer composition of poly(acrylic acid)/sodium salt of carboxymethyl starch - Part I. Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 135, 529-535.
- Halder, A., Dhall, A. and Datta, A. K. (2007). An improved, easily implementable, porous media based model for deep-fat frying. Part I: Model development and input parameters. *Food and Bioproducts Processing* 85, 2009-219.
- Higuchi, T. (1961). Rate of release of medicaments from ointment bases containing drugs in suspension. *Journal of Pharmaceutical Sciences* 50, 874-875.
- Higuchi, T. (1963). Theoretical analysis of rate of release of solid drugs dispersed in solid matrices. *Journal of Pharmaceutical Sciences* 52, 1145-1149.
- Kochhar, P. and Gertz, C. (2004). New theoretical and practical aspects of the frying process. *European Journal of Lipid Science and Technology 106*, 722-727.
- Korsmeyer, R. W. and Peppas, N. A. (1981). Effect of the morphology of hydrophilic polymeric matrices on the diffusion and release of water soluble drugs. *Journal of Membrane Science 9*, 211-227.
- Krokida, M. K., Oreopoulou, V. and Maroulis, Z. B. (2000). Water loss and oil uptake as a function of frying time. *Journal of Food Engineering* 44, 39-46.
- Maitra, J. and Shukla, V. K. (2014). Cross-linking in hydrogels - A review. *American Journal of Polymer Science* 4, 25-31.

- Mandema, W. and Zeldenrust, H. (1977). Diffusion of polystyrene in tetrahidrofuran. *Polymer 18*, 835-839.
- Mohan, R. V. N., Delaney, R. A. and Mermelstein, N. (1995). An engineering perspective on deep-fat frying of breaded chicken pieces: Engineering aspects of deep-fat frying of foods. *Food Technology* 49, 138-141.
- Moreira, G. R. and Barrufet, M. A. (1996). Spatial distribution of oil after deep-fat frying of tortilla chips from a stochastic model. *Journal of Food Engineering* 27, 279-290.
- Moreira, R. G., Palau. J. and Sun, X. (1995). Simultaneous heat and mass transfer during the deep fat frying of tortilla chips. *Journal of Food Process Engineering 18*, 307-320.
- Moreira, R. G., Sun, X. and Chen, Y. (1997). Factors affecting oil uptake in tortilla chips in deep-fat frying. *Journal of Food Engineering 31*, 485-498.
- Moreno-Castro, L. E., Quintero-Ramos, A., Ruiz-Gutierrez, M. C., Sanchez-Madrigal, M. A., Melendez-Pizarro, C. O., Perez-Reyes, I. and Lardizabal-Gutierrez, D. (2015). Nixtamalization assisted with ultrasound: Effect on mass transfer and physicochemical properties of nixtamal, masa and tortilla. *Revista Mexicana de Ingeniería Química 14*, 265-279.
- Moussa, I. S. and Cartilier, L. H. (1996). Characterization of moving fronts in crosslinked amylose matrices by image analysis. *Journal of Controlled Release 42*, 47-55. Ni, H. and Datta, A. K. (1999). Moisture oil and energy transport during deep-fat frying of food materials. *Food and Bioproducts Processing 77*, 194-204.
- Olguin-Rojas, J. A., Vazquez-Leon, L. A., Salgado-Cervantes, M. A., Fernandez Barbero, G., Garcia-Alvarada, M. A. and Rodriguez-Jimenes, G. C. (2019). Water and phytochemicals dynamic during drying of red habanero chili pepper (*Capsicum Chinese*) slices. *Revista Mexicana de Ingeniería Química* 18, 851-864.
- Peppas, N. A. (1985). Analysis of Fickian and no-Fickian drug release from polymers. *Journal of Pharmaceutical Sciences* 60, 110-111.

- Ritger, P. L. and Peppas, N. A. (1987a). A simple equation for description of solute release I. Fickian and non-Fickian release from nonswellable devices in the form slabs, spheres, cylinders or discs. *Journal of Controlled Release* 5, 23-36.
- Ritger, P. L. and Peppas, N. A. (1987b). A simple equation for description of solution release II. Fickian and anomalous release from swellable devices. *Journal of Controlled Release* 5, 37-42.
- Rubnov, M. and Saguy, I. S. (1997). Fractal analysis and crust water diffusivity of a restructured potato product during deep-fat frying. *Journal* of Food Science 62, 135-154.
- Santiago-Ramos, D., Figueroa-Cárdenas, J. F. C., Mariscal-Moreno, R. M., Escalante-Aburto, A., Ponce-García, N. and Véles-Medina J. J. (2018). Physical and chemical changes undergone by pericarp and endosperm during corn nixtamalization-A review. *Journal of Cereal Science 81*, 108-117.

- Siepmann, J. and Peppas, N. A. (2011). Higuchi equation: derivation, applications, use and misuse. *International Journal of Pharmaceutics* 418, 6-12.
- Soppirnath, K. S., and Aminabhavi, T. M. (2002). Water transport and drug release study from cross-linked polyacrylamide grafted guar gum hydrogel microspheres for the controlled release application. *European Journal of Pharmaceutics and Biopharmaceutics 53*, 87-98.
- Stier, R. F. (2004). Frying as a science an introduction. *European Journal of Lipid Science* and Technology 106, 715-721
- Tomasula, P. M. and Craig, J. C. (1991). Correlation of moisture and oil concentration in French fries. *Lebensmittel-Wissenschaft* + *Technology* 24, 445-448.
- Tong, C. H. and Lund, D. B. (1990). Effective moisture diffusivity in porous materials as a function of temperature and moisture content. *Biotechnology Progress* 6, 67-75.