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Modeling of kinetics, equilibrium and distribution data of osmotically dehydrated carambola (*Averrhoa carambola* L.) in sugar solutions

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ABSTRACT

Osmotic dehydration (OD) of carambola slices in sucrose, fructose and glucose solutions has been carried out to evaluate water and solute diffusivities, as well as the final impregnation–dehydration levels of the fruit. OD kinetics was performed in sugar solutions (50 g/100 g) at 45, 60 and 75 °C during 10 h using a syrup-to-fruit mass ratio of 15:1. An analytical solution for unsteady-state mass transfer based on Fick's second law of diffusion was used for the mathematical description of water loss and solute gain kinetics. By following a central composite design, additional OD tests were conducted to evaluate the effect of solute concentration (35.9–64.1 g solute/100 g solution) and process temperature (38.8–81.2 °C) on the equilibrium and distribution data for both solutes and water. Under the described experimental conditions, effective water diffusivity was in the range of $1.00–3.74 \times 10^{-9}$ m^2/s, whereas values for sucrose, fructose and glucose diffusivities were between $0.58–1.79 \times 10^{-9}$, $0.56–1.34 \times 10^{-9}$ and $0.56–1.88 \times 10^{-9}$ m^2/s, respectively. Results demonstrated that sucrose can be considered a better osmotic agent than fructose and glucose for OD of carambola, favoring greater water loss-to-solute gain ratios at comparable mass transfer rates.

1. Introduction

Starfruit or carambola (*Averrhoa carambola* L.) is a popular fruit cultivated in tropical countries. This fruit is usually consumed fresh or made into juice or juice drinks (Liew-Abdullah et al., 2007), and represents an important source of natural antioxidants such as vitamin C, carotenoids, and some phenolics compounds (Shui and Leong, 2004, 2006). In addition, carambola has been reported to be a rich source of insoluble dietary fiber with potential hypoglycemic effects (Chau et al., 2004). Carambola is a highly perishable fruit because of its high moisture content which can lead to extensive postharvest losses caused by chemical and microbial deterioration. Thus, the development of simple and inexpensive processes for its preservation is desirable (Karim and Wai, 1999). Existing studies have focused on the effect of chemical agents and modified atmospheres on shelf life extension of fresh-cut carambola (Teixeira et al., 2007, 2008). A feasible way of increasing shelf life of carambola is through the development of reduced moisture products. However, even though air drying is the most widely used technique for dehydrated fruit and vegetable production, it may be detrimental, affecting both nutritional and sensory quality of heat-sensitive products (Ruiz-López et al., 2010). Moreover, available information on dehydration processes of carambola is limited. Selected works include those from Karim and Wai (1999) which studied foam-mat drying characteristics of carambola. Recently, Chen et al. (2010) assessed the drying performance of an experimental solar energy-assisted photocatalytic low-pressure dryer using carambola as test sample. Osmotic dehydration (OD) is a processing technology which can be applied to produce partially dehydrated carambola with increased potential for product development and wider marketability.

OD is a mass transfer operation where water is partially removed from foods by immersing them in concentrated aqueous solutions such as syrups or brines. Because cell membranes function as semi-permeable films, water loss is accompanied by a simultaneous solute gain from the solution into the product. This operation has gained a broad acceptance in fruits and vegetable processing as it improves quality characteristics such as color, texture and flavor in the final product (Bidaisee and Badrie, 2001; Pereira et al., 2006). Moreover, since OD is able to produce a significant dewatering of foods, energy requirements for a further thermal drying can be considerably reduced (Ruiz-López et al., 2010). A good understanding of mass transfer rates for both solute gain and water loss as well as the final impregnation–dehydration levels of a

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Nomenclature

b_0,b_1,b_2,b_{12},b_{11},b_{22}	fitted parameters for second order linear model
D	effective diffusivity (m^2/s)
L	characteristic length for diffusion (m)
m	mass (kg)
SG	solute gain (kg solids/kg fresh product)
t	time (s)
WL	water loss (kg water/kg fresh product)
X	mass fraction (wet basis) of a given component in food product
x_1	coded concentration in second order linear model
x_2	coded temperature in second order linear model
y	fitted response in second order linear model
Y	mass fraction (wet basis) of a given component in osmotic solution

Greek symbols

λ	equilibrium distribution coefficient

Subscripts

0	at the beginning of the OD process
i	represents the i-th component distributed between the continuous (osmotic solution) and solid (product) phases
p	for product
s	for solute
t	at time t
w	for water
∞	at equilibrium

given product under different OD conditions (such as osmotic agent concentration, process temperature and solute type) is required for an adequate design of this operation (Shi and Le Maguer, 2002).

Mass transfer phenomena occurring during OD of foodstuffs have been frequently described by analytical or numerical solutions to Fick's second law of diffusion for several shapes and boundary conditions, where mass transfer rates for both water loss and solute gain are characterized by their effective diffusivities (Li and Ramaswamy, 2006; İspir and Toğrul, 2009; Rózek et al., 2009; Allali et al., 2010; Corrêa et al., 2010; Monnerat et al., 2010). These models have been successfully applied to estimate water and solute diffusivities during OD of several fruits and vegetables in syrups, brines or sugar–salt mixtures (İspir and Toğrul, 2009; Monnerat et al., 2010; Mercali et al., 2011). On the other hand, the study of equilibrium and distribution data for water and solutes is necessary to determine the final impregnation–dehydration levels of a food product under specific process conditions. Distribution coefficients have been successfully modeled as a function of both solute concentration and process temperature during OD of vegetables such as pineapple, potato, apple, mango and apricot (Parjoko et al., 1996; Rahman et al., 2001; Sablani et al. 2002; Sablani and Rahman, 2003; Toğrul and İspir, 2008) in sugar solutions. The same approach has been followed during the modeling of distribution coefficients in osmodehydrated sardine sheets using brines as osmotic solutions (Corzo and Bracho, 2004).

Nowadays, no information regarding the mass transfer rates for water loss and solute gain as well as the final impregnation–dehydration levels during OD of carambola is available. Therefore, the objective of this work was to study the kinetics, equilibrium and distribution data during OD of carambola slices in sucrose, fructose and glucose syrups.

2. Materials and methods

2.1. Raw material

Fresh well-graded carambola fruits (*Averroa carambola* L. cv. Golden star) were supplied by a local producer from Tuxtepec, Oaxaca (México). The maturity level of carambola fruits was evaluated according to the color classification given by Abdullah et al. (2006). In this study, fruits with a maturity level 3 were only selected, as they have been considered to render the best quality products. This maturity level corresponds to *ripe* category (25–75% trace of yellow in the skin color).

2.2. Total soluble solids (TSS)-to-total titratable acidity (TTA) ratio

In addition, total soluble solids (TSS)-to-total titratable acidity (TTA) ratio was determined to further characterize the maturity level of the fruits as recommended by Narain et al. (2001). TSS was determined by means of a refractometer (Atago model Master-T, Tokyo, Japan) as described by the AOAC method 983.17 (AOAC, 1999). TSS by refractometric method is defined as the concentration (by weight) of sucrose in solution having the same refractive index as the analyzed solution, and is generally expressed as degrees Brix (Bx), i.e., g sucrose/100 g solution. On the other hand, TTA, expressed in grams of anhydrous citric acid per 100 g of sample (g a.c.a./100 g product), was evaluated according to AOAC method 983.17 (AOAC, 1999). All analyses were carried out by triplicate using the mixed juice of three fruits in each determination.

2.3. Osmotic dehydration kinetics

Carambola slices (1 cm thickness) were subjected to duplicate OD processes in glucose, fructose or sucrose solutions (50 g/100 g) at 45, 60 and 75 °C during 10 h using magnetic stirring at about 120 rpm. On average 4, slices were obtained from each fruit as the tips were discarded. A syrup-to-fruit mass ratio of 15:1 was used in all experiments to avoid a significant change in syrup concentration (García et al., 2007). The slices were suspended in a wire mesh basket which was placed into air-tight glass jars filled with the osmotic solution. Samples were removed from the solution (without replacement) at regular intervals (every 20 min during the first 3 h, every 30 min during the next 2 h, every 40 min during the following 2 h, and every 60 min from hour 7 onward), quickly rinsed with distilled water, gently blotted dry with a paper towel to remove adhering osmotic solution, and then analyzed for their moisture and solute contents (X_w and X_s, respectively) to calculate water loss (WL) and solute gain (SG).

2.4. Equilibrium and distribution data for water and solute during osmotic dehydration

A second set of experiments was conducted to characterize the effect of temperature and osmotic media concentration on the equilibrium and distribution data of solutes and water during OD of carambola. Test conditions were obtained from a central composite design and included a total of nine duplicated treatments (18 experiments) with five different levels of each factor for every solute previously used. This design was implemented to obtain a set of conditions covering the experimental factor space in such way that fitted models had uniform prediction capabilities in that region (since all experimental points are distributed in a symmetrical and regular way for both factors and equidistant to the center of the factor space). Table 1 shows the range and levels of variables investigated. In these experiments, carambola slices were immersed in syrups for a total of 24 h. This time was considered as enough for carambola slices to reach mass equilibrium within the proposed solutions according to previous studies conducted with other plant materials (Rahman et al.,2001; Sablani et al. 2002; Sablani and Rahman, 2003). Experimental setup and remaining processing conditions and procedures are the same as those described in the previous section. Water loss and solute gain at equilibrium (WL_∞ and SG_∞, respectively) were obtained as the response variables in these experiments.

2.5. Water loss and solute gain calculations

Fresh and osmotically dehydrated samples were oven dried until constant mass weight was attained (when mass change was less than 0.001 g over an 8 h period) to determine their moisture and solute contents. These data were further used to evaluate water loss and solute gain with the following equations (see Nomenclature section for variable definitions):

$$WL_t = \frac{m_{po}X_{wo} - m_{pt}X_{wt}}{m_{po}} \tag{1}$$

$$SG_t = \frac{m_{pt}X_{st} - m_{p0}X_{s0}}{m_{p0}} \tag{2}$$

2.6. Equilibrium and distribution data calculations

A stationary state for both WL and SG kinetics is reached if OD is performed for a sufficiently long time. Under such circumstances, the net mass transfer rate between food and osmotic solution is zero. This period can be characterized by equilibrium and distribution coefficients of the diffused substances between involved phases. An adequate understanding of these variables is required for OD process modeling as a unit operation and for setting the required impregnation–dehydration levels of product. An equilibrium distribution coefficient for the i-th component between the continuous (osmotic solution) and solid (product) phases can be defined as,

$$\lambda_{i\infty} = \frac{X_{i\infty}}{Y_{i\infty}} \tag{3}$$

Eq. (3) can be written in terms of the initial concentration of the osmotic solution instead of its equilibrium concentration (Rahman et al., 2001; Sablani et al. 2002; Sablani and Rahman, 2003; Corzo and Bracho, 2004; Toğrul and İspir, 2008), providing that syrup-to-fruit mass ratio is very large (i.e., the concentration of the osmotic solution remains constant along the process). Thus, the equilibrium distribution coefficient for water can be defined as,

$$\lambda_{w\infty} = \frac{X_{w\infty}}{Y_{w0}} \tag{4}$$

Table 1
Central composite design levels for studying equilibrium and distribution data of osmotically dehydrated carambola slices.

Variables	Real and coded levels[*]				
Solute concentration (g/100 g)[**]	35.9 ($-\alpha$)	40 (-1)	50 (0)	60 (1)	64.1 (α)
Temperature (°C)	38.8 ($-\alpha$)	45 (-1)	60 (0)	75 (1)	81.2 (α)

[*] $\alpha = (2^k)^{1/4}$, where k is the number of studied factors ($k = 2$), thus $\alpha \approx 1.4142$.

[**] Sucrose, fructose or glucose.

Analogously, the equilibrium distribution for solute is written as,

$$\lambda_{s\infty} = \frac{X_{s\infty}}{Y_{s0}} = \frac{1 - X_{w\infty}}{1 - Y_{w0}} = \frac{1 - Y_{w0}\lambda_{w\infty}}{1 - Y_{w0}} \tag{5}$$

Since both product and osmotic solution can be considered as pseudo-binary systems, the following useful relationships can be defined,

$$X_{w\infty} = \frac{X_{w0} - WL_\infty}{1 - WL_\infty + SG_\infty} \tag{6}$$

$$X_{s\infty} = \frac{X_{s0} + SG_\infty}{1 - WL_\infty + SG_\infty} \tag{7}$$

2.7. Modeling of the osmotic dehydration kinetics

Analytical solutions for one-dimensional mass transfer by diffusion within a flat slab were used to describe OD kinetics (Crank, 1975),

$$WL_t = WL_\infty\left\{1 - \frac{8}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{(2n-1)^2} exp\left[\frac{-(2n-1)^2\pi^2 D_w t}{4L^2}\right]\right\} \tag{8}$$

$$SG_t = SG_\infty\left\{1 - \frac{8}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{(2n-1)^2} exp\left[\frac{-(2n-1)^2\pi^2 D_s t}{4L^2}\right]\right\} \tag{9}$$

where the following assumptions are made during their development: (i) constant concentration of the osmotic solution, (ii) negligible resistance to mass transfer at product surface, and (iii) isothermal process. Similar solutions are available for several geometries and have been used extensively to represent the mass transfer kinetics during OD of several fruits and vegetables (Li and Ramaswamy, 2006; İspir and Toğrul, 2009; Rózek et al., 2009; Allali et al., 2010; Corrêa et al., 2010).

Eqs. (3) and (4) were fitted to experimental data using non-linear regression to estimate water and solute diffusivities (D_w and D_s), as well as equilibrium dehydration and solute impregnation levels of the product (WL_∞ and SG_∞). Water and solute diffusivities were further used to calculate the dehydration efficiency index, defined as the water-to-solute diffusivities ratio (D_w/D_s) (Lazarides et al., 1997). The fitness quality of the proposed models was assessed by the determination coefficient (R^2), whereas significance of the non-linear regression parameters was tested by constructing their 95% confidence intervals (95% CI). In addition, analysis of variance was conducted on the estimated diffusivities and dehydration efficiency indices to evaluate their dependency on temperature and solute type. Non-linear regression and statistical analysis were performed with the Matlab Statistics Toolbox 5.0 (MathWorks Inc., Natick, MA, USA).

2.8. Modeling of the equilibrium and distribution data

Equilibrium (WL_∞, SG_∞, WL_∞/SG_∞ and $X_{w\infty}$) and distribution data ($\lambda_{w\infty}$ and $\lambda_{s\infty}$) were related to process variables by a second order model with main, interaction and quadratic terms, which is able to describe possible curvature of the responses:

$$Y = b_0 + b_1x_1 + b_2x_2 + b_{12}x_1x_2 + b_{11}x_1^2 + b_{22}x_2^2 \tag{10}$$

where x_1 and x_2 represent the coded levels for the osmotic media concentration and process temperature, respectively. Eq. (9) was fitted to experimental data using linear regression to estimate model parameters b_0, b_1, b_2, b_{12}, b_{11} and b_{22} for each one of the responses studied. The fitness quality of the linear regression models was assessed by the determination coefficient (R^2). Following data fitting, an analysis of variance of each model was performed to identify the significant predictor variables ($p = 0.05$). Linear regression and statistical analysis were performed with the Matlab Statistics Toolbox 5.0 (MathWorks Inc., Natick, MA, USA).

3. Results and discussion

3.1. Raw material

TSS, TTA, and TSS-to-TTA ratio were determined as 9.13 ± 1.27°Bx, 0.77 ± 0.18 g a.c.a./100 g product, and 12.23 ± 3.02°Bx/(g a.c.a./100 g product), respectively (mean ± s.d.). In addition, moisture content of fresh carambola was evaluated as 92.14 ± 1.04 g water/100 g product (mean ± s.d.). The values for moisture content and TSS-to-TTA ratio obtained in this work are similar to those reported by Narain et al. (2001) for carambola growing in Paraíba, Brazil, from the same cultivar and maturity level [90.32 ± 0.98 g water/100 g product and 14.31 ± 2.99°Bx/(g a.c.a./100 g product), mean ± s.d.].

3.2. Osmotic dehydration kinetics

Summarized statistics for the regression analysis of water loss and solute gain kinetics are shown in Tables 2 and 3, respectively. As it can be observed from the 95% CI, the non-linear regression analysis produced significant parameter estimates for all OD data ($p < 0.05$). Experimental and predicted water loss data are plotted in Fig. 1 and the corresponding results for solute gain are plotted in Fig. 2. In all cases, a good agreement was found between experimental and predicted results ($R^2 > 0.89$). The experimental variability observed in Figs 1 and 2 is mainly due to the fact that every kinetic was performed with slices obtained from several carambola fruits. In fact, even when the maturity level was controlled there were small (but appreciable) differences in color, sweetness and texture between fruits. It is important to mention that even when final dehydration level (WL_∞) for sucrose and fructose at 60 °C appears slightly above that of the kinetic conducted at 75 °C, both values cannot be considered statistically different (Table 2). Water diffusivities were in the range of 1.00–3.74 × 10^{-9} m^2/s, while values for sucrose, fructose and glucose diffusivities were between 0.58–1.79 × 10^{-9} m^2/s, 0.56–1.37 × 10^{-9} m^2/s and 0.56–1.88 × 10^{-9} m^2/s, respectively. Effective diffusion coefficients of solutes were lower than those of water

Table 2
Estimated water diffusivity and water loss at equilibrium during osmotic dehydration of carambola slices*.

Solute and temperature	$D_w \times 10^9$ (m^2/s)	WL_∞ (g/g)	R^2
Sucrose, 45 °C	1.0498 (0.8871/1.2125)	0.7087 (0.6779/0.7396)	0.9665
Sucrose, 60 °C	2.2081 (1.9867/2.4294)	0.7098 (0.6971/0.7226)	0.9770
Sucrose, 75 °C	2.7818 (2.4897/3.0739)	0.6946 (0.6827/0.7066)	0.9739
Fructose, 45 °C	0.9973 (0.8447/1.1500)	0.6075 (0.5800/0.6350)	0.9681
Fructose, 60 °C	2.4202 (2.1202/2.7202)	0.6118 (0.5975/0.6262)	0.9661
Fructose, 75 °C	3.3029 (2.8410/3.7648)	0.5912 (0.5781/0.6043)	0.9599
Glucose, 45 °C	1.1343 (0.9708/1.2977)	0.6762 (0.6498/0.7025)	0.9613
Glucose, 60 °C	2.6663 (2.4216/2.9111)	0.6662 (0.6553/0.6771)	0.9825
Glucose, 75 °C	3.7407 (3.3853/4.0960)	0.6802 (0.6705/0.6898)	0.9826

* Values in parentheses indicate the 95% CI.

Table 3
Estimated solute diffusivity and solute gain at equilibrium during osmotic dehydration of carambola slices*.

Solute and temperature	$D_s \times 10^9$ (m^2/s)	SG_∞ (g/g)	R^2
Sucrose, 45 °C	0.5784 (0.2550/0.9019)	0.0710 (0.0554/0.0866)	0.8957
Sucrose, 60 °C	1.4882 (1.1551/1.8213)	0.0757 (0.0719/0.0795)	0.8943
Sucrose, 75 °C	1.7865 (1.4212/2.1519)	0.0768 (0.0733/0.0804)	0.9237
Fructose, 45 °C	0.5596 (0.3371/0.7822)	0.1072 (0.0908/0.1236)	0.9314
Fructose, 60 °C	1.2610 (1.0305/1.4916)	0.1112 (0.1058/0.1167)	0.9476
Fructose, 75 °C	1.3371 (1.1021/1.5722)	0.1224 (0.1168/0.1280)	0.9443
Glucose, 45 °C	0.5617 (0.3060/0.8173)	0.1085 (0.0893/0.1277)	0.8954
Glucose, 60 °C	1.2891 (1.0150/1.5633)	0.1110 (0.1045/0.1176)	0.9185
Glucose, 75 °C	1.8806 (1.6185/2.1428)	0.1170 (0.1133/0.1206)	0.9621

* Values in parentheses indicate the 95% CI.

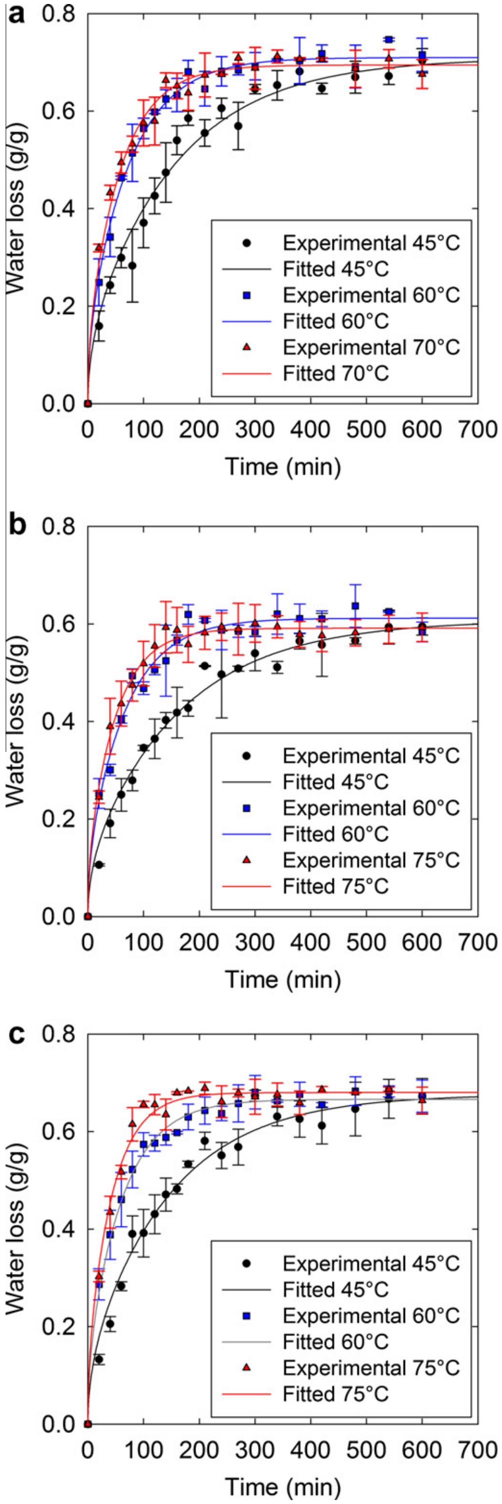

Fig. 1. Experimental and predicted water loss during osmotic dehydration of carambola slices with (a) sucrose, (b) fructose and (c) glucose solutions.

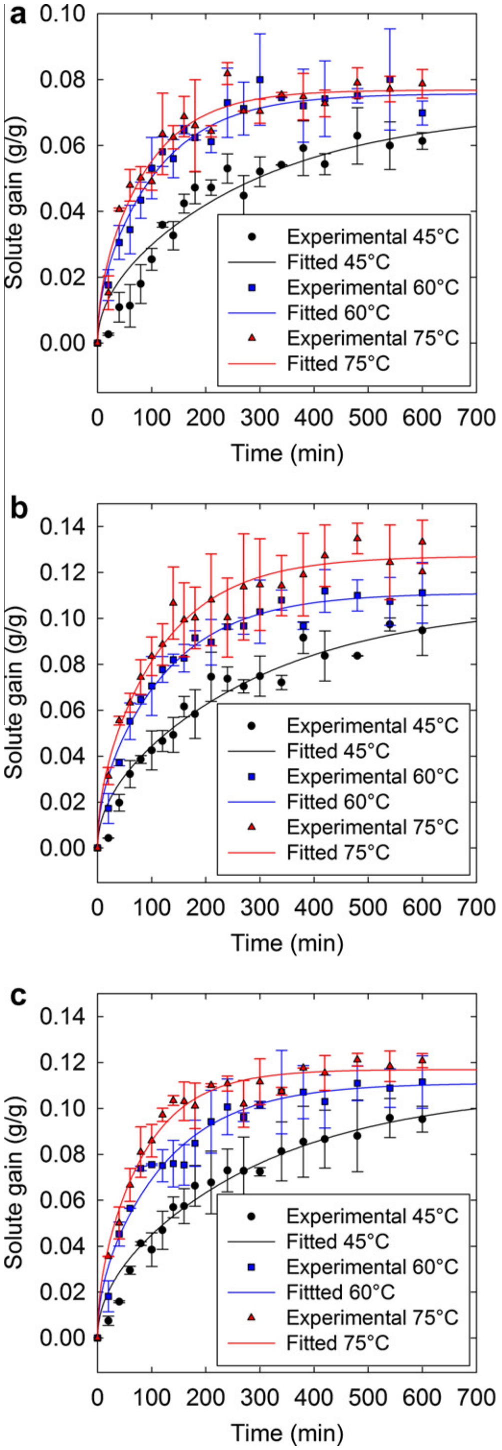

Fig. 2. Experimental and predicted solute gain during osmotic dehydration of carambola slices with (a) sucrose, (b) fructose and (c) glucose solutions.

for all treatments. As expected, the use of higher processing temperatures increased diffusion coefficients for both water and solutes ($p < 0.05$). On the other hand, solute type had an effect on estimated water diffusivities, but solute diffusivities exhibited negligible differences between sugars ($p < 0.05$). Additionally, the dehydration efficiency index was comparable for all solutes and was found to be independent on solute type and temperature ($p < 0.05$). The dehydration efficiency indices for sucrose, fructose and glucose were calculated as 1.62 ± 0.17, 2.06 ± 0.36, and 2.02 ± 0.04, respectively (mean ± s.d.). It is important to mention that color was the most significant change in product quality. Carambola slices processed at 60 °C developed a dark yellow color after 10 h of OD for all sugars. However, the color of carambola slices processed at 75 °C ranged from light orange to light brown. Further changes in color were observed during equilibrium experiments under the same OD conditions because of their longer processing times.

Fructose and glucose (molecular weight = 180.155) would be expected to exhibit higher diffusivities than sucrose (molecular weight = 342.296) because of their smaller molecular size theoretically allowing a greater mobility in food media (Lazarides et al., 1997; Kowalska et al., 2008). Differences in molecular weight also cause that sucrose solutions have a greater water activity and lower osmotic pressure (i.e., lower driving potential for mass transfer) than fructose and glucose syrups of the same mass concentration because of its corresponding lower molar concentration (Atarés et al., 2008). However, all solutes showed similar diffusivities. Thus, solute diffusivity results cannot be explained in terms of the molecular weights of sugars. Sucrose solutions have the greatest viscosities at all temperatures and mass concentrations in comparison with fructose or glucose syrups (Telis et al., 2007). Solution viscosity might be able to explain why the lowest water diffusivities were estimated when sucrose was used as osmotic agent (an increased external resistance to mass transfer). Nevertheless, this theory cannot explain observed differences in water diffusivity for fructose and glucose (fructose syrups have lower viscosities than glucose solutions but water diffusivity was higher when glucose was used as solute). Therefore, results suggest that additional chemical or physical interactions between food and solute may be occurring during OD process (Mercali et al., 2011).

The effect of solute type on mass transfer rates for water loss and solute gain has been addressed by some authors, but different conclusions have been described depending on product and experimental procedures. Atarés et al. (2008) observed higher mass transfer rates for water loss and solute gain during pulsed vacuum OD of apple cylinders (30 °C) when sucrose was used as osmotic agent in comparison with glucose (using osmotic solutions with the same water activity and thus leading to glucose syrups with reduced mass concentration, 30 g/100 g *versus* 41 g/100 g). However, Kowalska et al. (2008) reported higher diffusivities for water and solute during OD of pumpkin cubes (30 °C) when glucose was used as osmotic agent in comparison with sucrose, even when glucose was used at a lower concentration (49.5 g/100 g *versus* 61.5 g/100 g). Recently, İspir and Toğrul (2009) conducted the OD of apricot with sucrose, fructose and glucose solutions (70 g/100 g, 25–45 °C). In this study, water and solute diffusivities with sucrose and fructose solutions were found to be similar and above the values calculated for glucose.

Estimated water and solute diffusivities during OD of carambola compare favorably with reported values for other fruits. For example, glucose and sucrose diffusivities during OD of pumpkin have been estimated as 0.97×10^{-9} m^2/s ($D_w/D_s = 1.90$) and 0.68×10^{-9} m^2/s ($D_w/D_s = 1.75$), respectively (Kowalska et al., 2008). Similar results have been obtained during OD of apple (34–66 °C) and mango (40–80 °C) in sucrose syrups, with solute diffusivities in the ranges of 0.78–3.72 × 10^{-9} m^2/s ($D_w/D_s = 0.82 \pm 0.26$, mean ± s.d.) and 0.47–1.15 × 10^{-9} m^2/s ($D_w/D_s = 1.30 \pm 0.17$, mean ± s.d.), respectively (Alakali et al., 2006; Li and Ramaswamy, 2006).

3.3. Equilibrium and distribution data

The proposed central composite design along with the response for each experiment is shown in Table 4. Missing values for $X_{w\infty}$ and WL_∞/SG_∞ can be easily obtained from the other variables. It should be noticed that values for $X_{w\infty}$, WL_∞ and SG_∞ can be used

Table 4
Equilibrium and distribution data for water and solute during osmotic dehydration of carambola slices*.

Run	Concentration (g/100 g)	Temperature (°C)	Sucrose				Fructose				Glucose			
			WL_∞	SG_∞	$\lambda_{w\infty}$	$\lambda_{s\infty}$	WL_∞	SG_∞	$\lambda_{w\infty}$	$\lambda_{s\infty}$	WL_∞	SG_∞	$\lambda_{w\infty}$	$\lambda_{s\infty}$
1	40 (−1)	45 (−1)	0.5982	0.0955	1.0533	0.9201	0.5297	0.1365	1.0951	0.8574	0.5746	0.1301	1.0335	0.9497
2	40 (−1)	45 (−1)	0.6309	0.0904	1.0460	0.9311	0.5546	0.1525	1.0949	0.8576	0.5798	0.1200	1.0291	0.9564
3	40 (−1)	75 (1)	0.5325	0.1309	1.0296	0.9556	0.6142	0.1340	1.0762	0.8857	0.5607	0.1670	1.0083	0.9875
4	40 (−1)	75 (1)	0.5376	0.1057	1.0397	0.9404	0.5377	0.1300	1.0645	0.9032	0.5497	0.1361	1.0523	0.9216
5	60 (1)	45 (−1)	0.7513	0.0474	1.3224	0.7851	0.7126	0.1246	1.2420	0.8387	0.7796	0.1095	1.0911	0.9393
6	60 (1)	45 (−1)	0.7772	0.0705	1.2459	0.8361	0.7304	0.1068	1.2782	0.8146	0.7774	0.0954	1.1906	0.8730
7	60 (1)	75 (1)	0.6991	0.1104	1.2326	0.8449	0.7442	0.1220	1.2351	0.8433	0.7698	0.1110	1.0843	0.9438
8	60 (1)	75 (1)	0.7192	0.0845	1.2245	0.8503	0.7239	0.1215	1.2398	0.8402	0.7561	0.1166	1.1169	0.9221
9	35.9 (−α)	60 (0)	0.5248	0.1161	1.0214	0.9618	0.4720	0.1526	1.0626	0.8879	0.4989	0.1593	1.0160	0.9715
10	35.9 (−α)	60 (0)	0.5516	0.1300	1.0113	0.9798	0.4548	0.1316	1.0740	0.8676	0.4958	0.1619	1.0109	0.9805
11	64.1 (α)	60 (0)	0.8001	0.0821	1.2746	0.8465	0.7516	0.1116	1.3427	0.8084	0.7855	0.1027	1.1491	0.9166
12	64.1 (α)	60 (0)	0.7942	0.0812	1.2642	0.8523	0.7577	0.1155	1.3440	0.8077	0.7832	0.1127	1.1869	0.8955
13	50 (0)	38.8 (−α)	0.7483	0.0636	1.1317	0.8683	0.6727	0.1098	1.1669	0.8331	0.7335	0.1071	1.0703	0.9297
14	50 (0)	38.8 (−α)	0.7397	0.0619	1.1506	0.8494	0.6375	0.1204	1.1253	0.8747	0.7200	0.1123	1.0705	0.9295
15	50 (0)	81.2 (α)	0.7121	0.1227	1.0078	0.9922	0.6532	0.1481	1.1244	0.8756	0.6917	0.1370	1.0225	0.9775
16	50 (0)	81.2 (α)	0.6705	0.1314	1.0808	0.9192	0.6238	0.1283	1.1729	0.8271	0.6868	0.1428	1.0403	0.9597
17	50 (0)	60 (0)	0.6624	0.1213	1.1300	0.8700	0.6305	0.1403	1.1355	0.8645	0.6959	0.1304	1.0473	0.9527
18	50 (0)	60 (0)	0.6682	0.0993	1.1104	0.8896	0.6594	0.1331	1.1034	0.8966	0.6704	0.1376	1.0613	0.9387

[*] Values in parentheses represent the coded levels for osmotic media concentration (x_1) and process temperature (x_2), respectively. $\alpha \approx 1.4142$.

with Eq. (5) to obtain the initial mass fraction of water in product X_{w0} reported in Section 3.1. In all cases, distribution coefficients of solutes were lower than those of water. Distribution coefficients for water were in the range of 1.01–1.32 with corresponding values for solute between 0.79–0.99 when sucrose was used as osmotic agent. Similar results were obtained during the OD of carambola in fructose ($\lambda_s = 0.81$–0.90, $\lambda_w = 1.06$–1.34,) and glucose ($\lambda_s = 0.87$–0.99, $\lambda_w = 1.01$–1.19) solutions. However, higher water loss-to-solute gain ratios (WL_∞/SG_∞) were obtained for osmotically dehydrated carambola in sucrose solutions (4.07–15.85), in comparison with those of fructose (3.09–6.89) or glucose (3.06–8.15) syrups. Regression model parameters for the investigated responses are shown in Table 5. Fitted models can be used to predict equilibrium and distribution data in the concentration and temperature ranges of 36–64 g/100 g and 39–81 °C, respectively. Analysis of variance of the estimated constants demonstrated that syrup concentration had a significant effect on all studied variables for each osmotic agent ($p < 0.05$). Moreover, all solutes exhibited identical trends in terms of the main effects. An increase of syrup concentration produced higher values in WL_∞, $\lambda_{w\infty}$ and WL_∞/SG_∞, though the opposite trend was observed for SG_∞, $\lambda_{s\infty}$ and $X_{w\infty}$. As the OD process continues, a stationary state is reached when the chemical potential of syrup and fruit become equal. Since water activity can be decreased both by WL or SG, there is an inverse relationship between these variables, i.e. if WL is higher, then SG must be lower, and vice versa (Parjoko et al., 1996). Moreover, at higher syrup concentrations, solids could accumulate on product surface, limiting a further solute uptake since higher rates for external mass transfer are obtained. On the other hand, the influence of temperature on the studied responses was different for sucrose, fructose and glucose. Temperature did not exhibit a significant effect on the equilibrium and distribution data when carambola was osmotically dehydrated in fructose solutions. However, the use of higher processing temperatures produced an increase of SG_∞, reducing both WL_∞ and WL_∞/SG_∞ when glucose or sucrose were used as osmotic agents ($p < 0.05$). This may be due to the decrease of syrup viscosity at higher temperature which affects the external mass transfer rate (Parjoko et al., 1996).

Table 5
Regression coefficients of response surface models for equilibrium and distribution data during osmotic dehydration of carambola slices*.

Solute	Response	Parameters						R^2
		b_0	b_1	b_2	b_{12}	b_{11}	b_{22}	
Sucrose	WL_∞	**0.6653**	**0.0863**	**−0.0261**	0.0061	−0.0080	0.0169	0.9365
	SG_∞	**0.1103**	**−0.0142**	**0.0193**	0.0033	−0.0057	−0.0094	0.8538
	$\lambda_{w\infty}$	**1.1202**	**0.0983**	**−0.0259**	−0.0102	0.0192	−0.0059	0.9322
	$\lambda_{s\infty}$	**0.8798**	**−0.0484**	**0.0245**	0.0036	0.0087	0.0073	0.8362
	WL_∞/SG_∞	**6.0949**	**2.1589**	**−2.1397**	**−0.9927**	0.5263	**1.3179**	0.9061
	$X_{w\infty}$	**0.5601**	**−0.0655**	**−0.0125**	−0.0033	0.0002	−0.0034	0.9638
Fructose	WL_∞	**0.6450**	**0.0937**	0.0029	−0.0053	−0.0141	0.0048	0.9423
	SG_∞	**0.1367**	**−0.0099**	0.0033	0.0046	−0.0041	−0.0047	0.6341
	$\lambda_{w\infty}$	**1.1195**	**0.0901**	−0.0055	0.0005	**0.0405**	0.0112	0.9641
	$\lambda_{s\infty}$	**0.8806**	**−0.0228**	0.0061	−0.0055	**−0.0170**	−0.0121	0.7176
	WL_∞/SG_∞	**4.7241**	**1.1200**	−0.1460	−0.2128	0.1283	0.2346	0.8991
	$X_{w\infty}$	**0.5597**	**−0.0735**	−0.0028	0.0014	0.0109	0.0060	0.9879
Glucose	WL_∞	**0.6832**	**0.1019**	**−0.0113**	0.0016	**−0.0226**	**0.0109**	0.9955
	SG_∞	**0.1340**	**−0.0169**	**0.0101**	−0.0038	−0.0015	−0.0062	0.8718
	$\lambda_{w\infty}$	**1.0543**	**0.0498**	−0.0121	−0.0098	0.0195	−0.0005	0.8540
	$\lambda_{s\infty}$	**0.9457**	**−0.0209**	0.0104	0.0063	−0.0044	−0.0004	0.6128
	WL_∞/SG_∞	**5.1044**	**1.4955**	**−0.5320**	0.0004	0.0913	**0.3821**	0.9620
	$X_{w\infty}$	**0.5272**	**−0.0834**	−0.0055	−0.0039	0.0045	<0.0001	0.9867

[*] Bold numbers indicate significant parameter estimates ($p < 0.05$).

Similarly, growing values for $\lambda_{w\infty}$ and $X_{w\infty}$ with the simultaneous lowering of $\lambda_{s\infty}$ were obtained for increasing temperatures, but only with sucrose syrups ($p < 0.05$). Representative plots for water and solute distribution coefficients obtained with fructose solutions are shown in Figs 3 and 4, respectively. The effect of temperature and osmotic solute concentration has been documented during the modeling of distribution data ($\lambda_{w\infty}$ and $\lambda_{s\infty}$) in other fruits and vegetables, with comparable behaviors (Rahman et al., 2001; Sablani et al. 2002; Sablani and Rahman, 2003; Toğrul and İspir, 2008).

Other studies (Parjoko et al., 1996; Rahman et al., 2001; Sablani et al. 2002; Sablani and Rahman, 2003) have reported comparable results for the distribution coefficients of both water and solute during OD of other food products such as pineapple ($\lambda_s = 0.80$–1.15, $\lambda_w = 0.29$–1.42), potato ($\lambda_s = 0.90 = 1.53$, $\lambda_w = 0.72$–1.05,), apple ($\lambda_s = 0.47$–1.49, $\lambda_w = 0.62$–1.80,) and mango ($\lambda_s = 0.52$–1.18, $\lambda_w = 0.91$–2.12) in sucrose solutions. Recently, Toğrul and İspir (2008) investigated the OD of whole apricots (40–70 g/100 g, 25–45 °C) with various osmotic agents (sucrose, fructose, glucose, maltodextrin and sorbitol) reporting water and solute values of water and solute distribution coefficients in the ranges of 0.72–1.47 and 0.65–1.30, respectively. Although similar values for water loss-to-solute gain ratios have been obtained during the OD of fruits such as kiwi ($WL_\infty/SG_\infty = 3.51$–$13.35$) and apple ($WL_\infty/SG_\infty = 3.88$–$7.24$) in sucrose syrups (Cao et al., 2006; Li and Ramaswamy 2006), limited studies comparing the proposed solutes have been published, with variable outcomes. For example, Nieto et al. (2004) found similar water loss-to-solute gain ratios during the OD of apple in sucrose and glucose syrups ($WL_\infty/SG_\infty \approx 4$), whereas Kowalska et al. (2008) reported higher values of WL_∞/SG_∞ during the OD of pumpkin when sucrose (9.35) was used as osmotic agent in comparison with glucose (2.32). In both cases, glucose was used in osmotic solutions with a lower mass concentration than sucrose. In contrast, a recent study has reported great differences between solutes during the OD of apricot with sucrose ($WL_\infty/SG_\infty = 36.63$–$54.38$), fructose ($WL_\infty/SG_\infty = 5.56$–$14.93$) and glucose ($WL_\infty/SG_\infty = 36.63$–$54.38$) syrups under the same process conditions (Toğrul and İspir, 2008).

Several authors have pointed out that total water removal can be predicted from the distribution coefficients and depending upon the need of keeping solute gain at a given level, syrup concentration and process temperature can be estimated (Rahman et al., 2001; Sablani et al., 2002; Sablani and Rahman, 2003). The same information can be obtained from the variables WL_∞/SG_∞ and $X_{w\infty}$ however, as shown in Table 5, these responses were generally fitted with higher determination coefficients and thus their use is recommended in this work. Moreover, the fitness quality of distribution data has been rather low in several fruits products such as pineapple [$R^2(\lambda_{w\infty}) = 0.48$, $R^2(\lambda_{w\infty}) = 0.68$], potato [$R^2(\lambda_{w\infty}) = 0.53$, $R^2(\lambda_{w\infty}) = 0.85$], apple [$R^2(\lambda_{w\infty}) = 0.77$, $R^2(\lambda_{w\infty}) = 0.83$] and mango [$R^2(\lambda_{w\infty}) = 0.71$, $R^2(\lambda_{w\infty}) = 0.73$] as reported in other studies (Rahman et al., 2001; Sablani et al., 2002; Sablani and Rahman, 2003). In fact, it should be considered that process differences in terms of distribution coefficients can be, indeed, enhanced or attenuated because their definitions, given by Eqs. (4) and (5), include the initial syrup concentration ($Y_{s0} = 1 - Y_{w0}$), which is also being used as

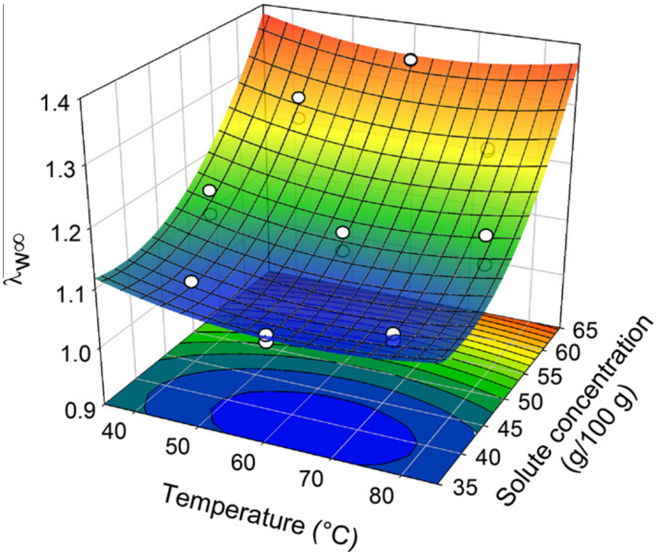

Fig. 3. Experimental and predicted water distribution coefficients for osmotically dehydrated carambola in fructose syrups.

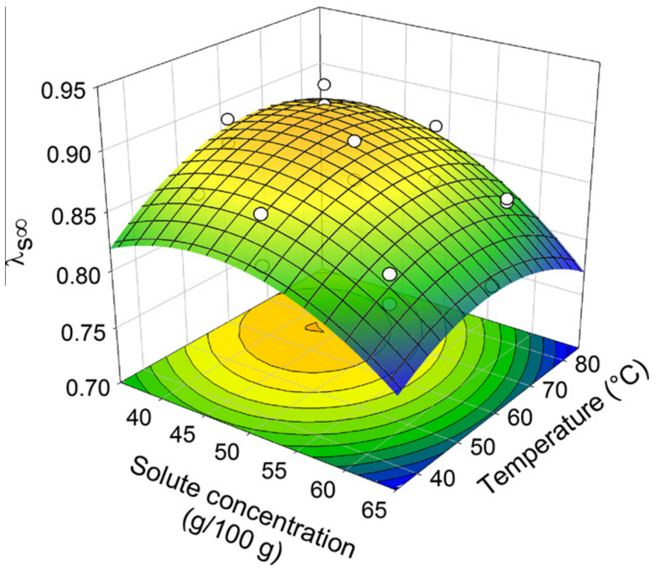

Fig. 4. Experimental and predicted solute distribution coefficients for osmotically dehydrated carambola in fructose syrups.

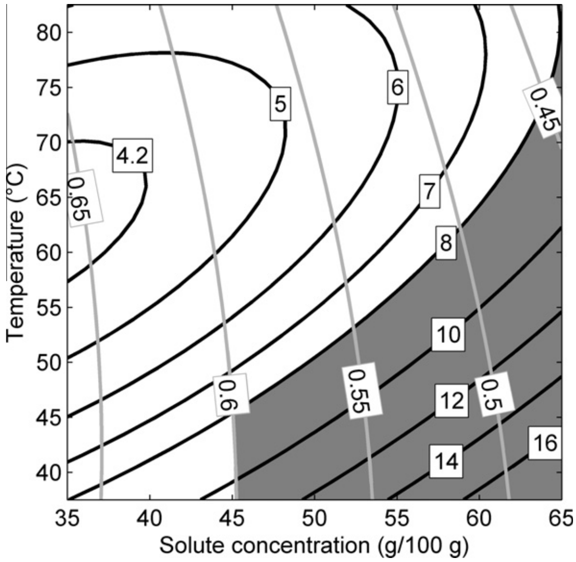

Fig. 5. Contour plots of water loss-to-solute gain ratio (WL_∞ SG_∞, black lines) and product moisture content ($X_{w\infty}$, gray lines) as a function of osmotic solution concentration and temperature for osmodehydrated carambola slices in sucrose solutions (gray region represents the set of conditions able to yield a product with $X_{w\infty} < 0.6$ g water/g product and WL_∞ $SG_\infty > 8$ g water/g solute).

a predictor variable (i.e., the variation source is included in response). Thus, process differences expressed in terms of the final water loss (WL_∞) and solute gain (SG_∞) or final water ($X_{w\infty}$) and solids ($X_{s\infty}$) content of the product become more relevant than actual differences in distribution coefficients. Figs 5–7 show the overlapped contour plots for water loss-to-solute gain ratio (WL_∞/SG_∞) and final moisture content of product ($X_{w\infty}$) as a function of syrup concentration (x_1) and temperature (x_2). These graphs can be used to determine the set of conditions yielding an osmodehydrated carambola product with specific final moisture content and solute impregnation level. For example, let us consider an OD process where the product must satisfy final moisture content lower than 0.6 g water/g product with a minimum water loss-to-solute gain ratio of 8 g water/g solute. The feasible set of process conditions is given by the gray region in Fig. 5 for osmotically dehydrated carambola in sucrose syrups. It should be noticed that even as fructose or glucose solutions can lower $X_{w\infty}$ up to the required level of 0.6 g water/g product, both solutes cannot achieve the desired $WL_\infty/SG_\infty > 8$ g water/g solute, resulting in a product with higher solute uptakes (Figs 6 and 7).

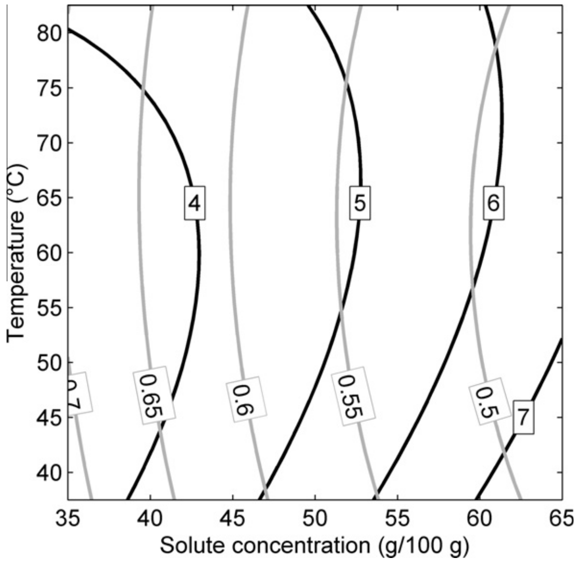

Fig. 6. Contour plots of water loss-to-solute gain ratio (WL_∞ SG_∞, black lines) and product moisture content ($X_{w\infty}$, gray lines) as a function of osmotic solution concentration and temperature for osmodehydrated carambola slices in fructose solutions.

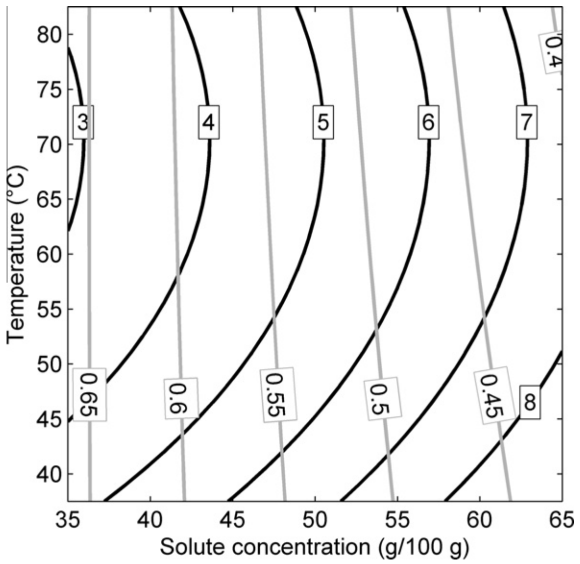

Fig. 7. Contour plots of water loss-to-solute gain ratio (WL_∞ SG_∞, R^2 black lines) and product moisture content ($X_{w\infty}$, gray lines) as a function of osmotic solution concentration (x_1) and temperature (x_2) for osmodehydrated carambola slices in glucose solutions.

4. Conclusions

Modeling of kinetics, equilibrium and distribution data during OD of carambola slices was performed to determine the water and solute diffusivities, as well as the final impregnation–dehydration levels of product. The dynamic period for mass transfer was described by the unsteady-state Fickian diffusion model, allowing the diffusion coefficients estimation, while effect of temperature and osmotic solute concentration on the experimental equilibrium and distribution data during the stationary period was characterized by second order response surface models. In this study, both water loss-to-solute gain ratio at equilibrium (WL_∞/SG_∞) and final moisture content of product ($X_{w\infty}$) were described with a higher precision than distribution coefficients for both water and solute. Thus, their combination is suggested as an option to characterize the equilibrium period during the OD of food products. From the mass transfer characteristics standpoint, results demonstrated that sucrose can be considered a better osmotic agent than fructose and glucose for OD of carambola, favoring greater water loss-to-solute gain ratios at comparable mass transfer rates. Further studies are needed to quantify the influence of solute type, temperature and syrup concentration on product characteristics related with consumer acceptance such as texture and color.

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