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Mass Transfer During Osmotic Dehydration of Chub Mackerel Cylinders in Ternary Solution

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Abstract

In the analysis, design and optimization of an osmotic dehydration process is important to know the kinetic of water loss and solutes gain. In this study, the mass transfer during osmotic dehydration of chub mackerel (*Scomber japonicus*) cylinders in ternary solution glycerol/salt/water was analyzed. The models of Zugarramurdi & Lupín and Azuara were used to describe mass transfer and to estimate equilibrium values. The radial effective diffusion coefficient was estimated using the analytical solution of Fick's second law. Diffusion coefficients were determined for a finite cylinder, for an infinite cylinder considering only the first term of the series and considering higher order terms of the series. The profiles of water and solutes during the osmotic dehydration were calculated by using the estimated water and solutes diffusivities. According to the results obtained, using three terms in the analytical solution of the Fick's second law is appropriate to determine the diffusion coefficients. The diffusion coefficient for infinite cylinder were 2.63×10^{-6}, 4.11×10^{-6} and 4.25×10^{-6} cm^2/s for water loss, salt and glycerol gain respectively. For a finite cylinder these values were 2.30×10^{-6}, 3.67×10^{-6} and 3.78×10^{-6} cm^2/s respectively. All the models proposed were in agreement with experimental data for solutes gain (($0.967<R^2_{adj}<0.986$); ($0.0016<RMSE<0.039$) and ($4.17<P<10.0$)). The model based on the solution of Fick's Law for an infinite cylinder with higher order terms was the best fit for water loss and solutes gain. The equilibrium values estimated with Azuara model agree with the experimental (0<relative error<9.8). Water and solute distributions as a function of time and location in the radial direction were plotted.

Keywords: chub mackerel, concentration profile, diffusion coefficient, mass transfer, osmotic dehydration

1. Introduction

Osmotic dehydration (OD) is a technique used for the partial removal of water when foods are placed in a hypertonic solution. During the process, two counter-current flows take place: water flows out from the food to the solution and the solute is transferred from the solution to the food. Mass transfer rate during osmotic dehydration depends on many factors such as concentration, temperature, composition of the osmotic solution, immersion time, nature of the food and their geometry, solution agitation, etc. (Li & Ramaswamy, 2006; Uribe et al., 2011; Abbasi Souraki, Ghaffari, & Bayat, 2012). For developing food-processing technology based on OD process, it is important to understand OD kinetics so as to set the desired dehydration level during the process. The kinetic of mass transfer can be predicted using an unsteady-state diffusion model (Fick's second law). Analytical solutions of the equation are available for idealized geometric media: spheres, infinite cylinders, infinite slabs, and semi-infinite media (Li & Ramaswamy, 2006) and the diffusion coefficients for both water loss and solids gain individually or simultaneously can be estimated. However, only limited research has been carried out on finite cylinders under typical osmotic dehydration conditions that describe mass transfer phenomena (Rastogi, Raghavarao, & Niranjan, 1997; Li & Ramaswamy, 2006; Abbasi Souraki et al., 2012). For fish products there is no information available on cylinder geometry.

Since diffusion equations have analytical solutions only for classical geometries, for non-classical geometries, numerical methods are necessary for their solution. Because of these limitations the use of empirical models is of interest, since these simple models have no geometric restrictions for their application. Water loss and solute gain kinetics during OD of fish products has been modeled by mean of empiric relations like Peleg and Zugarramurdi & Lupín models (Zugarramurdi & Lupín, 1980; Turhan, Sayar, & Gunasekara, 2002; Corzo & Bracho, 2005, 2006; Schmidt, Carciofi, & Laurindo, 2009; Czerner & Yeannes, 2010) and probabilistic models like Weibull (Corzo & Bracho, 2008). The information in the literature about the application of such empirical and probabilistic models for describing OD of fish products in ternary solutions containing glycerol is very scarce (Checmarev, Casales, Yeannes, & Bevilacqua, 2013; Checmarev, Casales, & Yeannes, 2013).

Chub mackerel (*Scomber japonicus*) is found along the Atlantic coast of South America. It is bounded on north by latitude 23°S (Rio de Janeiro) and on the south by latitude 39°S (Bahía Blanca) (Angelescu, 1980). In Argentine, during 2010 and 2011 the fishing fleet reached 27.558 and 28.253 tons respectively (MAGYP, 2011). The usual preservation of chub mackerel consists mainly in the traditional process of frozen, canned and smoked. Considering the aforementioned, it is important to develop new processing alternatives for this species. Osmotic dehydration represents a good alternative for the development of new products of chub mackerel.

In this work the kinetics of water loss and solid gain for chub mackerel cylinders during osmotic dehydration in water/salt/glycerol were studied. The empirical models of Zugarramurdi and Lupín and Azuara and a model based on the solution of Fick's Law for cylinder geometry were used. The water and solutes distributions during dehydration were predicted.

2. Materials and Methods

2.1 Raw Materials

Chub mackerel (*Scomber japonicus*) caught in Mar del Plata, Argentine, in October 2012 and stored at -18 °C was used in this study.

2.2 Process

The process of chub mackerel (*Scomber japonicus*) osmotically dehydrated consisted of the following stages: headed, gutted and tail cutting, steam cooking for 20 min, cooled at 7±1 ºC, obtainment of white muscle (cylindrical loin), immersion in water-salt-glycerol solution at 7±1 °C, drained off at 7±1 ºC, vacuum packed in flexible packaging and pasteurization (60±1 ºC).

2.3 Infusion Solution

Infusion solution composition (w/w) was: 55% water, 40% glycerol (Biopack, Zárate, Argentina, 99.5 g/100 g of purity), and 5% sodium chloride (Biopack, 99 g/100 g of purity). The a_w of infusion solution was 0.64. Relationship fish: solution was 1:10 (w/w) to avoid significant dilution of the osmotic solution as a result of water loss and solute uptake.

2.4 Sampling

Experimental works were carried out with long cylindrical samples of mackerel, so that the process could be considered as one-dimensional diffusion in the radial direction.

To determine the mass transfer kinetic and the diffusion coefficients, cylinders (cooked mackerel loin) of 16.5 cm long and 1.9 cm in diameter were immersed in ternary solution containing salt, water and glycerol and were kept under refrigeration temperature (7±1 °C). At different times, three cylinders were removed from the solution and the ends of the cylinders were discarded in order to prevent edge effects, only the central area of each cylinder (9 cm in length) were analyzed (Graiver, Pinotti, Califano, & Zaritzky, 2006). The contents of water, salt and glycerol in the samples were determined at 0.5, 1, 1.5, 2, 2.5, 3, 4, 7, 10.5, 15, 17, 19 and 24 h. The cylinders were drained, superficially rinsed with distilled water, dried with absorbent paper and weighed. Two experiments were carried out.

To validate the models were carried out two experiments in which immersion in the ternary solution was performed at 10 °C.

2.5 Physicochemical Analyses

The water content was determined at 105 °C until constant weight (AOAC, 1990) using a drying oven (Marne, 644, Córdoba, Argentine). The ashes were determined by calcinations at 550 °C as described by AOAC (1990) using an electric oven (Indef, 332, Córdoba, Argentine) and the sodium chloride content using the Mohr method adapted for food (Kirk, Sawyer, & Egan, 1996). The glycerol content was determined using an enzymatic UV method (Boehringer Mannheim/R-Biopharm) with a spectrophotometer (Shimadzu® UV-1601 PC, Kyoto, Japan). All analyses were done in triplicate.

2.6 Estimation of Effective Water, Glycerol and Salt Diffusivities

2.6.1 Mass Transfer Model Based on Fick's Second Law

The unsteady state Fickian diffusion model was applied to describe the water and solute diffusion into a cylinder (Crank, 1975).

$$\frac{\partial C}{\partial t} = D_e \frac{1}{r}\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial C}{\partial r}\right) \tag{1}$$

Analytical solution of Equation (1) is obtained by using the following initial and boundary conditions:

Uniform initial moisture and salt concentrations:

$$C(r,0) = C0 \tag{2}$$

Symmetry of concentration at the center:

$$\frac{\partial \mathrm{C}}{\partial \mathrm{r}} = 0 \text{ at } r = 0 \tag{3}$$

It is assumed that the shape and the diffusion coefficient are constant during dehydration. Constant equilibrium moisture and salt concentrations at the surface (negligible external resistance to mass transfer) are considered:

$$\mathrm{C(r, t)} = \mathrm{C_{eq}} \text{ for } r = R \tag{4}$$

Constant solution concentration (high solution to fish mass ratio).

Equation (1) and boundary conditions can be rewritten in dimensionless form:

$$\frac{\partial \varphi}{\partial \tau} = \frac{1}{x}\frac{\partial}{\partial x}\left(x\frac{\partial \varphi}{\partial \tau}\right) \varphi = \varphi(x,\tau);\ 0 \le x \le 1;\ \tau \ge 0 \tag{5}$$

$$\Phi\ (x,0)=1 \tag{6}$$

$$\frac{\partial \varphi}{\partial x} = 0 \quad x=0,\ \tau > 0 \tag{7}$$

$$\varphi(x,\tau) = 0 \quad x=0,\ \tau > 0 \tag{8}$$

Being

$$\varphi = \frac{C - C_{eq}}{C_0 - C_{eq}}\ ;\ \ x = \frac{r}{R}\ ;\ \tau = \frac{Dt}{R^2} \tag{9}$$

The solution of the Equation (5) for the average ratio of water loss and solute gain is given by:

$$\frac{x_t - x_{eq}}{x_0 - x_{eq}} = \sum \frac{4}{\delta_n^2} exp\left(-\delta_n^2 \frac{Dt}{R^2}\right) \tag{10}$$

Where x_t is the water loss or solute gain (expressed as g on a non-salt and non-glycerol dry matter basis, g/gdm) at time *t*, x_{eq} the equilibrium values and x_0 the initial values. The δ_n are the roots of the Bessel equation of the first kind of zero order: $Jo(\delta_n)=0$.

2.6.1.1 Model Based on Fick's Second Law for an Infinite Cylinder

The effective diffusion coefficient was calculated using 1, 3 and 5 terms of the Equation (10).

Equation (10) can be simplified by considering only the first term in their series expansion and written in logarithmic form as:

$$\ln\left(\frac{x_t - x_{eq}}{x_0 - x_{eq}}\right) = ln\frac{4}{\partial_1^2} - \delta_1^2 \frac{Dt}{R^2} \tag{11}$$

The water and solute effective diffusivities are derived from the slope of the plot of $ln\left(\frac{x_t - x_{eq}}{x_0 - x_{eq}}\right)$ versus t.

2.6.1.2 Model Based on Fick's Second Law for a Finite Cylinder

Prediction of mass transfer in a finite cylinder requires the use of analytical solutions obtained for both infinite cylinder and infinite slab. The solution of the Equation (5) for the average ratio of water loss and solute gain is given by:

$$\frac{x_t - x_{eq}}{x_0 - x_{eq}} = \left[\sum_{n=1}^{n=\infty} \frac{4}{\delta_n^2} exp\left(-\delta_n^2 \frac{Dt}{R^2}\right)\right]\left[\sum_{n=0}^{n=\infty} \frac{8}{(2n+1)^2\pi^2} exp\left(-\frac{(2n+1)^2\pi^2 Dt}{4l^2}\right)\right] \quad (12)$$

The effective diffusion coefficient was calculated using 5 terms of each series.

2.6.2 Water and Solute Profiles Into Mackerel Cylinder During Osmotic Dehydration

The concentration distribution of water and solutes is considered only along its radius r as a function of time t since the mackerel cylinder length l is many times larger than its diameter $2R$. The water and solutes distributions along the radius will be of the form (Crank, 1975):

$$\frac{C_{(r,t)} - C_{eq}}{C_0 - C_{eq}} = \sum_{n=1}^{\infty} \frac{2}{\delta_n J_1(\delta_n)} J_0(\delta_n \frac{r}{R}) \delta xp\left(-\delta_n^2 \frac{Dt}{R^2}\right) \quad (13)$$

or in the following dimensionless form:

$$\varphi(x,\tau) = \sum_{n=1}^{\infty} \frac{2}{\delta_n J_1(\delta_n)} J_0(\delta_n x) \exp\left(-\delta_n^2 \tau\right) \quad (14)$$

2.7 Empirical Models

Simple empirical models that have no geometric restrictions for their application have been reported for describing mass transfer in food subjected to OD and to predict water loss and solid gain at equilibrium condition (Azuara, Beristain, & García, 1992; Corzo & Bracho, 2006; Schmidt et al., 2009, among others).

2.7.1 Azuara Model

Azuara et al. (1992) developed a two parameters model with the following expression:

$$x_t = \frac{Stx_{eq}}{1+St} \quad (15)$$

where x_t and x_{eq} are water loss, salt or glycerol gain (expressed as g on a non-salt and non-glycerol dry matter basis, g/gdm) at time t and at equilibrium, respectively, and S (h^{-1}) is the model constant (rate constant).

2.7.2 Zugarramurdi and Lupin Model (Z&L Model)

Zugarramurdi and Lupín (1980) proposed a mathematical model, with an exponential approach to the salt and water equilibrium values:

$$\frac{\partial x_t}{\partial t} = k\left(x_{eq} - x_t\right) \quad (16)$$

where x_t and x_{eq} are the water loss, salt or glycerol gain (g/gdm) at a given time t (h) and at the equilibrium respectively and k is the specific rate constant (h^{-1}). Integrating Equation (16) with the initial $x_{t=0} = x_0$ condition is obtained:

$$x_t = x_0 e^{-kt} + x_{eq}(1 - e^{-kt}) \quad (17)$$

2.8 Statistical Analysis

The fitting of the models to the experimental data was performed using OriginPro 8. The goodness of fit was determined using the adjusted determination coefficient (R^2_{adj}), the root mean square error (*RMSE*, Equation (18)) and the average relative deviation (*P*, Equation (19)).

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(x_i - x_{pi}\right)^2} \quad (18)$$

$$P = \frac{100}{n}\sum_{i=1}^{n}\frac{|x_i - x_{pi}|}{x_i} \quad (19)$$

where x_i is the experimental value, x_{pi} is the predicted value and n the number of data pairs. Values of P less than or equal to 10% are considered to fit the experimental data satisfactorily (Azoubel & Murr, 2003).

3. Results and Discussion

3.1 Effective Diffusivities of Water and Solutes in Mackerel Cylinders

Effective diffusivities were calculated by fitting the unsteady Fick´s second law solutions (Equations (10), (11) and (12)) to the experimental data. The predicted values of water, salt and glycerol effective diffusivities for an infinite cylinder with 1, 3 and 5 terms of the series and for a finite cylinder are shown in Table 1. The values of R^2_{adj}, *RMSE* and *P* are shown in Table 2.

Table 1. Diffusion coefficients of water and solutes

Fick´s Model	$Dx10^6(cm^2/s)$ Infinite cylinder (5 terms of series)	Infinite cylinder (3 terms of series)	Infinite cylinder (linearized)	Finite cylinder
Water loss	2.63±0.28	2.63±0.30	2.88±0.50	2.30±0.25
Salt gain	4.14±0.11	4.11±0.14	4.04±0.31	3.67±0.15
Glycerol gain	4.25±0.34	4.25±0.36	4.06±0.40	3.78±0.33

Values are mean±standard deviation of two experiments.

Table 2. Statistical parameters of models

Fick´s Model	Infinite cylinder (5 terms of series)			Infinite cylinder (3 terms of series)			Infinite cylinder (linearized)			Finite cylinder		
	R^2_{adj}	RMSE	P(%)	R^2_{adj}	RMSE	P(%)	R^2_{adj}	RMSE	P(%)	R^2_{adj}	RMSE	P(%)
Water loss	0.952	0.045	8.46	0.947	0.048	8.88	0.930	0.18	14.90	0.924	0.057	10.0
Salt gain	0.980	0.030	4.17	0.976	0.032	4.35	0.979	0.12	8.75	0.967	0.039	5.69
Glycerol gain	0.987	0.023	4.32	0.985	0.024	4.48	0.986	0.10	7.82	0.984	0.025	4.34

In all cases, R^2_{adj} was higher than 0.924 and *RMSE* was lower than 0.18 indicating the acceptability of the model proposed. Values of *P* were less than or equal to 10%, excepting for diffusion coefficient of water loss estimated by the linearized equation. These results indicated that the models fit the experimental data. There were no significant differences to the 95% of confidence level between using three and five terms in the analytical solutions of the Fick second law to determine the diffusion coefficient. Therefore, an increase in the number of terms in the series did not generate changes in the diffusion coefficient as these terms were very small and negligible for the calculation. Ramallo, Schvezov, & Mascheroni (2004) studied the osmotic dehydration of pineapple in sucrose solution and reported that there were no significant differences between using three and four terms in the analytical solutions of Fick second law. When the diffusion coefficients were estimated by the linearized equation the *RMSE* was higher ($0.10<RMSE<0.18$) and also the *P* value for water loss was greater than 10%. Validation of models is shown in Figure 1, as can be seen the model followed the general trend of the experimental osmotic dehydration curves. The values of water loss, salt and glycerol gain predicted by the Fick model for infinite cylinder and for finite cylinder were very close to the experimental values. The model based on the solution of Fick's Law for an infinite cylinder was the best fit for water loss and salt gain. For glycerol gain there was no difference in the $R^2_{adj,}$ *RMSE* and *P* values of the model based on the solution of Fick´s Law for infinite and finite cylinder respectively.

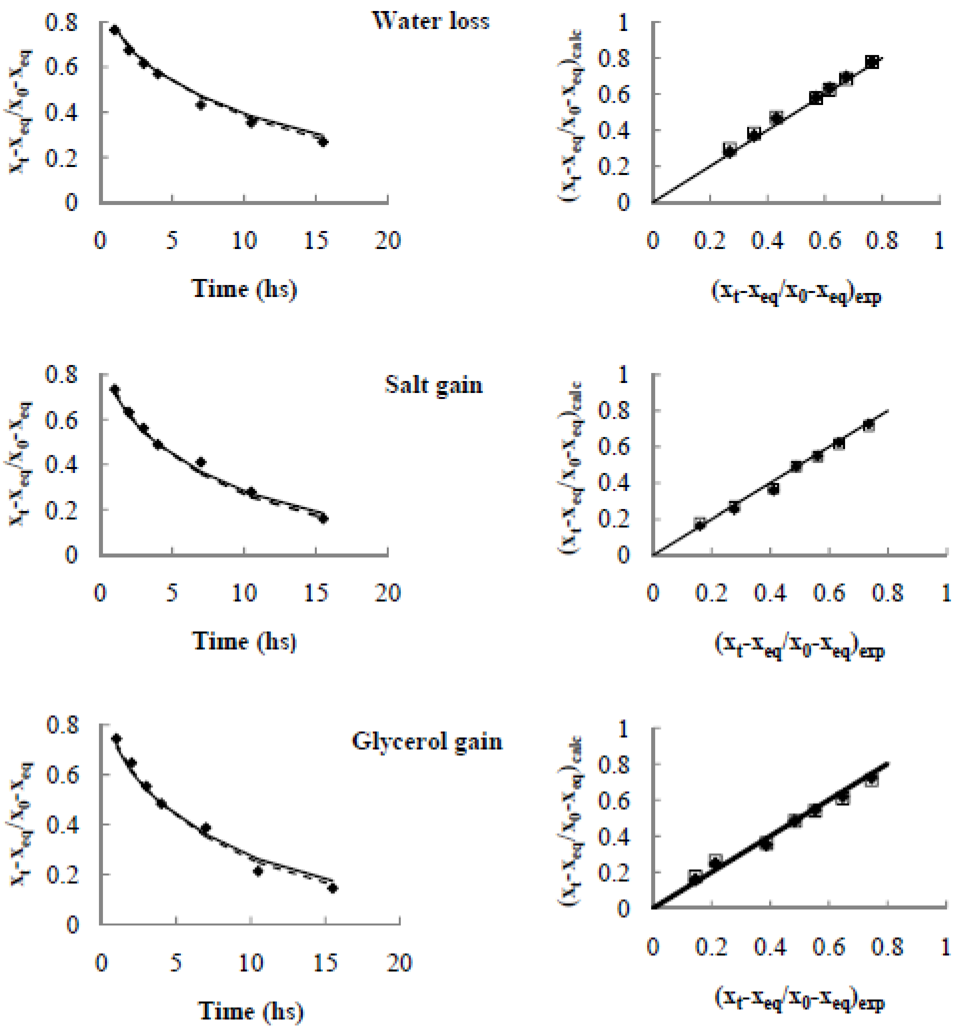

Figure 1. Fit of experimental data to Fick´s Models

3.2 Water and Solutes Profiles During Osmotic Dehydration

Since the predicted values of average water loss and solutes gain were found to fit well to the experimental data (Figure 1), the water, salt and glycerol distributions into mackerel cylinder were predicted. Equation (14) was solved using the predicted effective water, salt and glycerol diffusivities (Table 1). Dimensionless water, salt and glycerol contents vs. dimensionless time ($\tau = Dt/R^2$) were plotted as a function of dimensionless radius ($x = r/R$) in Figure 2. This Figure shows that the central point has the maximum water content and minimum salt and glycerol content (for water $C_o > C_{eq}$, for salt and glycerol $C_0=0$ and $C_0 < C_{eq}$). Therefore, water is transferred from interior to the surface and salt and glycerol are transferred into the sample in the reverse direction. As it can be seen in this figure, the decrease of water content and the increase of salt and glycerol contents is mainly carried out in the region of surface of the mackerel cylinder and slowly progresses to the interior. At the end of dehydration, the water, salt and glycerol contents in mackerel cylinder reached the equilibrium with osmotic solution.

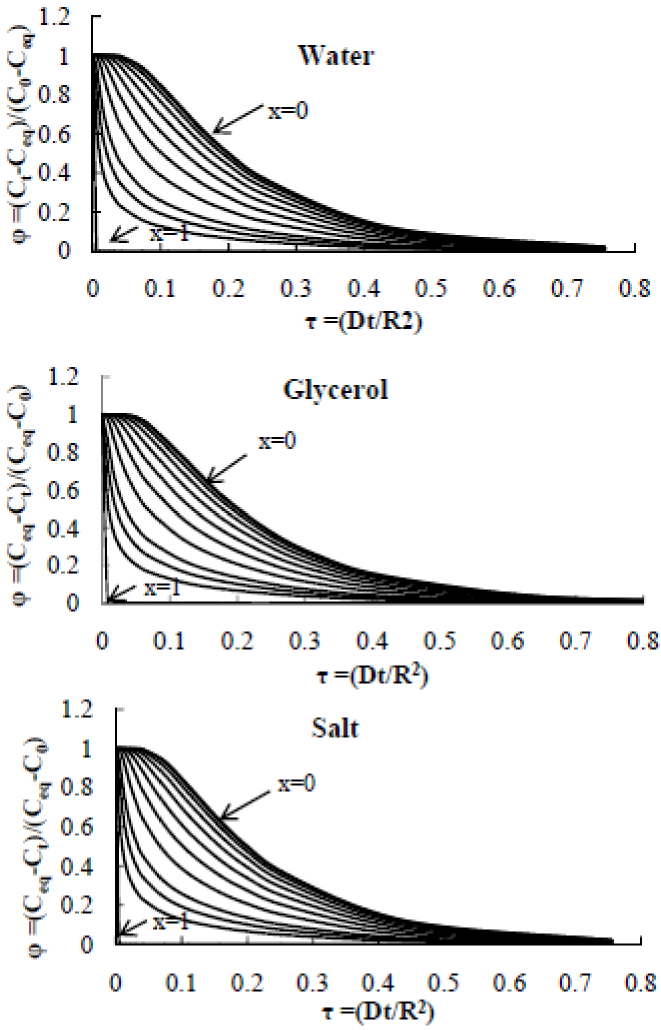

Figure 2. Dimensionless water, salt and glycerol distribution inside mackerel cylinder during osmotic dehydration

3.3 Kinetics of Osmotic Dehydration and Equilibrium Water Loss and Solutes Gain

Empirical models can be used to represent mass transfer during osmotic treatment, when the diffusive model is difficult to use or when experimental determination of the equilibrium values requires long immersion times that can lead to food tissue changes. The models developed by Z&L and Azuara (Equations (15) and (17)) were used to predict the kinetics of osmotic dehydration and to determine the final water loss and solutes gain at equilibrium. The main advantage of these models is its capacity to predict the equilibrium values. Table 3 shows the parameters of the models as well as the R^2_{adj}, the root mean square error (*RMSE*) and *P* value. For solutes gain, in all cases the R^2_{adj} was higher than 0.93, *RMSE* was lower than 0.016 and *P* was less than or equal to 10 indicating the acceptability of the Z&L and Azuara models. For water loss, *P* value was greater than 10.

Table 3. Parameters of Z&L and Azuara models

	Z&L model					Azuara model				
	k (h^{-1})	x_{eq}	R^2_{adj}	RMSE	P(%)	S (h^{-1})	x_{eq}	R^2_{adj}	RMSE	P(%)
Water loss	0.186	0.451	0.972	0.018	11.81	0.1548	0.615	0.976	0.017	10.62
Salt gain	0.298	0.055	0.970	0.0024	7.88	0.2920	0.069	0.986	0.0016	5.56
Glycerol gain	0.342	0.385	0.931	0.024	10.00	0.3584	0.471	0.971	0.016	6.01

Validation of models is shown in Figure 3, as it can be seen, the water loss as well as solutes gain increased non-linearly with time and the predicted values are very close to the experimental values. Comparison of experimental and predicted equilibrium values is shown in Table 4. As can be seen in this table, Z&L model underestimates the equilibrium of water, salt and glycerol contents. On the other hand, the equilibrium values predicted by the Azuara model agree with the experimental ones according with the lower values of E.

Table 4. Comparison of experimental and predicted equilibrium values

	$x_{eqExp.}$	$x_{eqZ\&L\ model}$	E	$x_{eq\ Azuara\ model}$	E
Water loss	0.560	0.451	19.46	0.615	9.82
Salt gain	0.069	0.055	20.29	0.069	0
Glycerol gain	0.503	0.381	24.25	0.471	6.36

$E = \left| \frac{x_i - x_{pi}}{x_i} \right| 100$ E: relative error, x_i experimental value and x_{pi} predicted value.

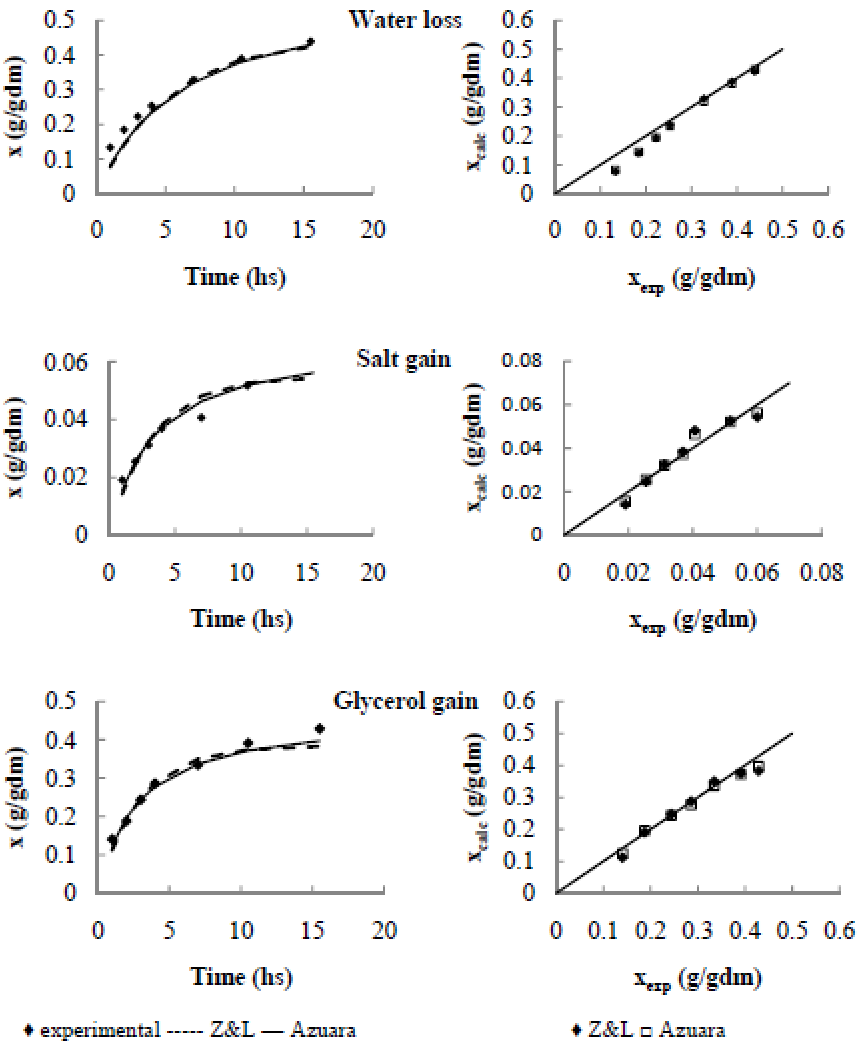

Figure 3. Fit of experimental data to Azuara and Z&L Models

4. Conclusions

In all the cases, Fick's Model for finite and infinite cylinder and empirical models of Z&L and Azuara better predicted the salt and glycerol gain than the water loss, during the osmotic dehydration of mackerel cylinders in hypertonic water/salt/glycerol solution.

Three terms in the analytical solution of the Fick equation for infinite cylinder is appropriate to determine the diffusion coefficients.

The model based on the solution of Fick's Law for an infinite cylinder was the best fit for water loss and solutes gain. The equilibrium values estimated with Azuara model agree with that determined experimentally.

Using calculated effective diffusivities of water, salt and glycerol, the water and solutes distributions into mackerel cylinder samples were predicted. The changes of water, glycerol and salt were mainly confined in the region of the surface and slowly progresses to the interior. At the end of the osmotic dehydration, the water, glycerol and salt in the mackerel cylinder reached the equilibrium with osmotic solution.

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