INVERSE PROBLEM ESTIMATING EFFECTIVE MASS DIFFUSIVITY CONSIDERING MASS TRANSFER DURING DRYING OF WEST INDIAN CHERRY

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Abstract. *This paper presents a comparative analysis between numerical the values of effective mass diffusivity of acerolas estimated by two optimization methods: Levenberg-Marquardt and Evolution Differential. Experimental results for moisture content within west Indian cherry during drying at air temperature 50°C were obtained for three cases: only convective drying, convective drying with previously 4 hours of osmotic dehydration, convective drying with previously 12 hours of osmotic dehydration, in order to fit proposed model of mass transfer. The effective diffusivity of west Indian cherry samples was estimated through of analytical solution of mass transfer equation, i.e., Fick´s 2nd law. The values obtained to effective mass diffusivity by two optimization methods have an excellent agreement between them and are in agreement with the values described of literature. The diffusivity values (by fitting the model to experimental data) were found to be in the range of* $10^{-11} \pm 10^{-12}$ *m^2/s.*

Keywords*: Levenberg-Marquardt, Evolution Differential, inverse problem, osmotic dehydration, convective drying.*

1. INTRODUCTION

Drying is one of the most important unit operations in the food process engineering, and represents a feasible way in order to extend the shelf life of foods with high moisture contents, especially fruits and vegetables, by reducing their water content to an extension at which the microbial spoilage and undesirable reactions are minimized. Additionally, drying of foodstuffs is intended to improve product stability, decrease shipping weights and costs and minimize packaging requirements. Because drying is an energy intensive operation, a better understanding of the drying mechanisms is important to optimize both the quality of the product and the efficiency of the process (Oliveira *et al*., 2006).

In recent years, due to the change in patterns of consumption and preference for minimally processed foods, has been given great attention to the transformation processes that preserve the physical structure and sensory characteristics of the products, mainly in products made from vegetables and fruits. Drying fruits is an alternative for obtaining improved products. The convective drying of foods (fruits) is a complex process which involves the simultaneous heat and mass transfer between the air and product. Apparently, in this process the phenomena are complex and involve understanding of fluid mechanics, thermodynamics, transfer heat, in addition to physical changes, chemical, and biochemical of the product.

The drying of foods using the heated air is based on increasing of food's temperature through of evaporation of the water, however if not well controlled, may causes undesirable changes in appearance, color, texture, flavour and nutrient content of the product (Shigematsu *et al.,* 2005).

A technique used to reduce the availability of water with minor damage to sensory and nutritional quality of foods is the osmotic dehydration. Osmotic dehydration is a water removal process, which is based on placing foods, such as fruits and vegetables, into concentrated solutions of soluble solids having higher osmotic pressure and lower water activity. Concentration results from simultaneous water and solute diffusion process caused by the water and solute activity gradients across the cell membrane. Since the membrane is only partially selective, there is always some solute diffusion into the food (Lima *et al.*, 2004).

According Cordova (2006), the osmotic dehydration followed by convective drying is a process efficient, producing fruits with better stability of color and texture. Dehydration reduces spoilage, increase shelf life, reduction of the product's mass and gives added value as it is without chemical treatments.

The inverse analysis is a technique efficient than use data for the estimation of constants that appears in mathematical models and to assist in the modeling of physical phenomenon. The interest in the analysis and solution of inverse problems of heat and mass transfer has been intensified recently. Many works of literature use deterministic or stochastic methods to minimize the objective function (Mendonça *et al*., 2005; Kanevce *et al*., 2005; Mariani and Coelho, 2006; Silva *et al*., 2006).

The deterministic methods are based on calculations of derived, with successive points in the space of optimization, requiring the search for knowledge of a vector direction of growth of function in case of problems of minimization, which depends on the gradient of function that will be minimized (Avila *et al.,* 2008). Classical deterministic methods classics are: maxima descent, Newton's method, Gradient, Levenberg-Marquardt, and simplex methods.

Modern optimization methodologies are being used to solve inverse problems, particularly stochastic methods, which usually supply potential solutions, but the computational time required by stochastic methods generally exceeds that of deterministic optimization methods.

The objective of this work is to provide a comparison between the values of effective mass diffusivity calculated by Levenberg-Marquardt and Evolution differential methods.

2. MATHEMATICAL FORMULATION

The developed model, based on Fick's unsteady state law of diffusion, calculates the amount of water leaving the fruit as a function of time, such equation is given below:

$$\frac{\partial X}{\partial t} = \nabla\left(D_{ef} \bullet \nabla X\right), \tag{1}$$

In spherical coordinates the equation (1) may be written in terms of r, θ and ψ, as:

$$\frac{\partial X}{\partial t} = \frac{1}{r^2}\left\{\frac{\partial}{\partial r}\left(D_{ef} r^2 \frac{\partial X}{\partial r}\right) + \frac{1}{\sin\theta}\frac{\partial}{\partial \theta}\left(D_{ef} \sin\frac{\partial X}{\partial \theta}\right) + \frac{D_{ef}}{\sin^2\theta}\frac{\partial^2 X}{\partial \psi^2}\right\} + \dot{n} \ , \tag{2}$$

Assuming that the mass transfer occurs only in the radial direction and there is no generation of mass, the equation (2) is reduced to the form:

$$\frac{\partial X}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(D_{ef} r^2 \frac{\partial X}{\partial r}\right), \tag{3}$$

Equation (3) should be also supplemented by the initial condition

$$X = X_0 \qquad t = 0, \qquad 0 < r < R \tag{4}$$

The following boundary conditions were assumed in problem discussed here, symmetry and equilibrium conditions, respectively (Park *et al.,* 2001; Brod *et al.,* 2003; Arévalo-Pinedo and Murr, 2005 and Oliveira *et al.,* 2006),

$$\frac{\partial X}{\partial r} = 0 \qquad t > 0, \qquad r = 0 \ , \tag{5}$$

$$X = X_{eq} \qquad t > 0, \qquad r = R \ , \tag{6}$$

Equation (3) whit the boundary conditions, equations (4) to (6), is solved analytically by separation of variables method. To solve the mass transfer equation the following assumptions were used:

- Effective mass diffusivity is constant;
- Just radial diffusion;
- The process is isothermal;
- Absence of generating mass;
- Homogeneous sample;
- Shrinkage is neglected.

The analytical solution of the equation (3) enables us to find the mass fraction of concentration in the west Indian cherry, given by

$$Y = \frac{\overline{X} - X_{eq}}{X_0 - X_{eq}} = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-n^2.\pi^2.D_{ef}.\frac{t}{r_1^2}\right), \tag{7}$$

where

Y:	dimensionless moisture content
$\bar{X}$:	average moisture content (kg water/ kg dry matter)
Xeq:	equilibrium moisture content (kg water/ kg dry matter)
X_0 :	initial moisture content (kg water/ kg dry matter)
D_{ef}:	effective mass diffusivity (m^2/s)
t:	time (s)
r_1:	average radius of the sample (m)

The equilibrium moisture content is obtained experimentally using the following equation,

$$X_{eq} = \frac{(m_{eq} - m_s)}{m_s}, \tag{8}$$

where m_{eq} is mass of the sample in the equilibrium, and m_s is dry mass of the sample.

3. MATERIAL AND METHODS

3.1. Materials

West Indian Cherry (Malpighia punicifolia L.) is a plant originated in Central America that has been propagated to South America including Brazil due to its good adaptation to soil and climate. It is nutritionally important, mainly due to its very high level of vitamin C, justifying the choice of this fruit to be studied in this work. The plant provides flowers and fruits at different stages and, consequently, long fruiting periods during the year are observed. The fruit presents a short living period after being picked (2 to 3 days), at room temperature. Ripening of the fruit involves a series of complex biochemical reactions such as hydrolysis of starch, conversion of chloroplasts into chromoplasts with chlorophyll transformation, production of carotenoids, anthocyanins and phenolics and the formation of volatile compounds. All these reactions are important for the characteristics of the mature fruit and for its peculiar flavour (Speirs and Brady, 1991; Vendramini and Trugo, 2000)

West Indian Cherry ripe, but firm, buy in supermarket of João Pessoa (city located in Paraiba – Brazil) were used as samples. Samples having a minimum diameter of 85mm were selected to ensure the assumption of constant average radius of the sample.

3.2. Experimental Procedure

A - Initially the fruits were selected, washed and dried with paper towels;
B - It was determined the radius and average initial weight of the three samples that were placed in aluminum container and brought to the drier for monitoring the weight loss, by 39 hours to a temperature of 50ºC;
C - After 1h of drying and each subsequent hour, made up the weighing of samples;
D - Following 39 hours of drying, the samples were taken to oven to 65 ° C for 24 hours for the determination of dry weight.

Procedures A→D were repeated for samples submitted to pré-treatment osmotic in a binary sucrose solution at 65° Brix, in proportion fruit: solution of 1:10 by 4h and 12h, respectively.

4. INVERSE PROBLEM

For the inverse problem of interest here, the effective mass diffusivity $D_{ef,}$ is regarded as unknown parameter while all other parameters used in the solution of the problem are known direct. To determine the effective mass diffusivity D_{ef}, we believe that D_{ef} is constant for a given temperature, moreover, are used experimental data of the evolution of transient moisture content (Y_i), when the product is subjected to a drying process with or without osmotic dehydration. The subscribed *i* refers to the time in which measurements were made (i = 1, ..., m). The estimation methodology used is based on the minimization of the ordinary least square norm.

$$\mathbf{S}(\mathbf{P}) = [\mathbf{Y} - \mathbf{X}(\mathbf{P})]^T [\mathbf{Y} - \mathbf{X}(\mathbf{P})], \tag{9}$$

where,

$\mathbf{P}[P_1, P_2.P_3..., P_N]$, is the vector of unknown parameters. $[\mathbf{Y}-\mathbf{X}(\mathbf{P})]^T$ is given by $[\mathbf{Y}-\mathbf{X}(\mathbf{P})]^T = [(\vec{Y}_1 - \vec{X}_1)(\vec{Y}_2 - \vec{X}_2)......(\vec{Y}_i - \vec{X}_i)]$, where $(Y_i - X_i)$ is the vector lines that contain the differences between the moisture content obtained experimentally in time you and he estimated from a theoretical model.

4.1. Levenberg-Marquardt Method

A version of the method of Levenberg-Marquardt was applied to solving the problem of reverse estimation of parameter. The solution for vector **P** is achieved using the following iterative procedure:

$$P^{r+1} = P^r + \left[(\mathbf{J}^r)^T \mathbf{J}^T + \mu \mathbf{I}\right]^{-1} (\mathbf{J}^r)^T \left[\mathbf{Y} - \mathbf{X}(\mathbf{P}^r)\right] \tag{10}$$

Where **J** is the sensitivity matrix defined as:

$$\mathbf{J}(\mathbf{P}) = \begin{bmatrix} \frac{\partial X_1^T}{\partial P_1} & \cdots & \frac{\partial X_1^T}{\partial P_N} \\ \cdot & \cdots & \cdots \\ \frac{\partial X_1^T}{\partial P_1} & \cdots & \frac{\partial X_1^T}{\partial P_N} \end{bmatrix} = \left[\frac{\partial X^T(\mathbf{P})}{\partial \mathbf{P}}\right] \tag{11}$$

The term μ I is a regulator of instability introduced due to bad conditioning characteristic of the problem. This prevents the matrix J^TJ is not natural at the beginning of repetition, therefore, the procedure is the slow convergence of the method of step downward. The iterative process is concluded that the standard of the gradient of **S**(**P**) is sufficiently small, or whether the change in the vector of parameters $\mathbf{P}^{r+1} - \mathbf{P}^r$ is very small (Press *et al.,* 1989)

4.2. Evolution Differential Method

Differential Evolution (DE) is a population-based stochastic function minimizer (or maximizer) relating to Evolutionary Algorithms, whose simple yet powerful and straightforward features make it very attractive for numerical optimization. DE combines simple arithmetical operators with the classical operators of recombination, mutation and selection to evolve from a randomly generated starting population to a final solution. DE uses mutation based on distribution of solutions in the current population (Storn and Price, 1995; 1997). In this way, search directions and possible step sizes depend on the location of the individuals selected to calculate the mutation values (Liu and Lampinen, 2002). It evolutes generation by generation until the termination conditions have been met.

The different variants of DE are classified using the following notation: DE/α/β/δ, where α indicates the method for selecting the parent chromosome that will form the base of the mutated vector, β indicates the number of difference vectors used to perturb the base chromosome, and δ indicates the recombination mechanism used to create the offspring population. The *bin* acronym indicates that the recombination is controlled by a series of independent binomial experiments.

The fundamental idea behind DE is a scheme whereby it generates the trial parameter vectors. In each step, the DE mutates vectors by adding weighted, random vector differentials to them. If the cost of the trial vector is better than that of the target, the target vector is replaced by the trial vector in the next generation. The variant implemented here was the DE/*rand*/1/*bin*, which involved the following steps and procedures:

Step 1: *Initialization of the parameter setup*: The user must choose the key parameters that control the DE, i.e., population size, boundary constraints of optimization variables, mutation factor (f_m), crossover rate (*CR*), and the stopping criterion (t_{max}).

Step 2: *Initialize the initial population of individuals*: Initialize a population of individuals (solution vectors) with random values generated according to a uniform probability distribution in the *n*-dimensional problem space.

Step 3: *Evaluate the objective function value*: For each individual, evaluate its objective function (fitness) value.

Step 4: *Mutation operation (or differential operation)*: Mutate individuals in according to equation:

$$z_i(t+1) = x_{i_1}(t) + f_m \cdot [x_{i_2}(t) - x_{i_3}(t)] \quad (12)$$

In the above equations, i =1,2,...,N is the individual's index of population; t is the time (generation); $x_i(t) = \left[x_{i_1}(t), x_{i_2}(t), ..., x_{i_n}(t)\right]^T$ stands for the position of the i-th individual of population of N real-valued n-dimensional vectors; $z_i(t) = \left[z_{i_1}(t), z_{i_2}(t), ..., z_{i_n}(t)\right]^T$ stands for the position of the i-th individual of a *mutant vector*; f_m > 0 is a real parameter, called *mutation factor*, which controls the amplification of the difference between two individuals so as to avoid search stagnation. The mutation operation randomly select the target vector $x_{i_1}(t)$, with $i \neq i_1$.Then, two individuals $x_{i_2}(t)$ and $x_{i_3}(t)$ are randomly selected with $i_1 \neq i_2 \neq i_3 \neq i$, and the difference vector x_{i_2} - x_{i_3} is calculated.

Step 5: *Crossover (recombination) operation*: Following the mutation operation, crossover is applied in the population. For each mutant vector, $z_i(t+1)$, an index $rnbr(i) \in \{1, 2, \cdots, n\}$ is randomly chosen using a uniform distribution, and a *trial vector*, $u_i(t+1) = \left[u_{i_1}(t+1), u_{i_2}(t+1), ..., u_{i_n}(t+1)\right]^T$, is generated with

$$u_{i_j}(t+1) = \begin{cases} z_{i_j}(t+1) \text{ if } randb(j) \leq CR \text{ or } j = rnbr(i), \\ x_{i_j}(t) \text{ otherwise,} \end{cases} \quad (13)$$

where *randb(j)* is the j-th evaluation of a uniform random number generation with [0, 1] and CR is a *crossover rate* in the range [0, 1].

To decide whether or not the vector $u_i(t + 1)$ should be a member of the population comprising the next generation, it is compared to the corresponding vector $x_i(t)$. Thus, if F_c denotes the objective function under minimization, then

$$x_i(t+1) = \begin{cases} u_i(t+1) \text{ if } f(u_i(t+1)) < f(x_i(t)), \\ x_i(t) \text{ otherwise,} \end{cases} \quad (14)$$

where f is the evaluation of cost function (objective function).

Step 6: *Verification of the stopping criterion*: Loop to Step 2 until a stopping criterion is met, usually a maximum number of iterations (generations), t_{max}.

5. RESULTS AND DISCUSSION

Three cases of drying were analyzed in this work, that follows,

Case 1: Only was made convective drying using 50°C at air temperature to dry acerolas by 39 hours.
Case 2: Convective drying was made using 50°C at air temperature to dry acerolas by 39 hours, previously was made osmotic dehydration by 4 hours using sucrose solution binary.
Case 3: Convective drying was made using 50°C at air temperature to dry acerolas by 39 hours, previously was made osmotic dehydration by 12 hours using sucrose solution binary.

In the table following are the values of effective mass diffusivity estimated for this work through the two methods of optimization.

Table 1. Values of effective mass diffusivity for different cases.

Optimization method	Levenberg-Marquardt	Differential Evolution
case 1	$8.104 \times 10^{-11} \pm 7.064 \times 10^{-12}$	8.60045×10^{-11}
case 2	$16.86 \times 10^{-11} \pm 38.69 \times 10^{-12}$	20.1264×10^{-11}
case 3	$9.554 \times 10^{-11} \pm 35.51 \times 10^{-12}$	12.6397×10^{-11}

Observing the table 1, realizes that the values of diffusivity estimated by the two methods present a wide agreement among themselves.

In figures 1 to 3, we will present the curves representing the temporal evolution of moisture, estimated by the two methods of optimization for the three cases investigated.

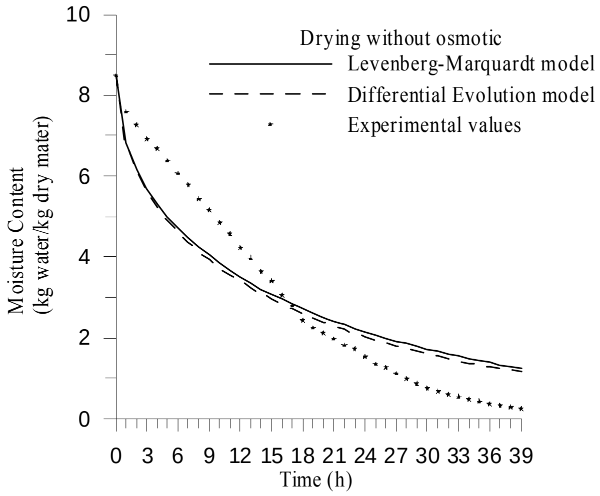

Figure 1. Models and experimental data for case 1.

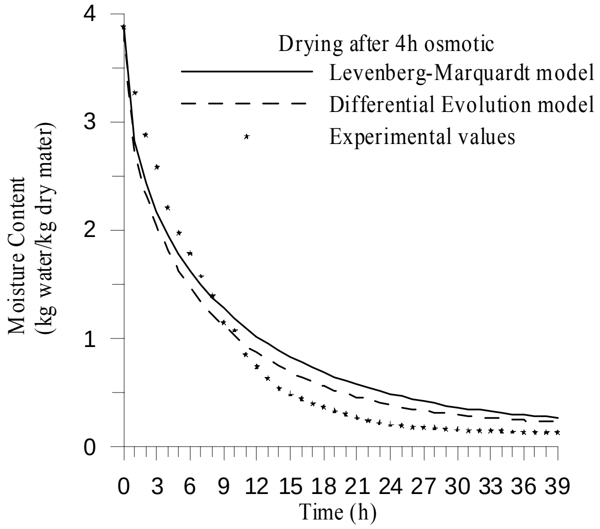

Figure 2. Models and experimental data for case 2.

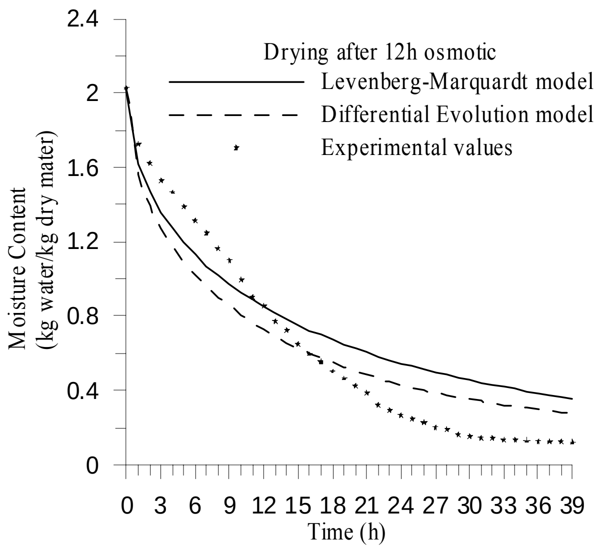

Figure 3. Models and experimental data for case 3.

In figures 1 to 3 we see that numerical results obtained by both the methods Levenberg-Marquardt and Differential Evolution are similar, however there are discrepancies when numerical results are compared with measured data, than can be explained by simplifications assumed in the solution of the Fick´s 2^{nd} law. It appears that the discrepancies shown in figure 1 are greater than shown in figures 2 and 3. This can be explained because of the potential for reducing the drying of the shell and resistance to mass transfer in fruit that is larger in nature than in osmotic dehydration they have suffered and also the shrinkage that occurs during drying is also higher in fruits that do not osmotic dehydration suffered earlier by the drying process.

In figure 3 note than the results obtained by optimization methods differ from measured data, larger than those observed in drying after 4 hours of osmotic dehydration, as presented in figure 2, this can be explained by the increased time that they were immersed in the solution osmotic, causing injuries on the fruit's surface, gain more solid and further reduction of the initial moisture.

In figures 4 to 6 are presented the curves representing the residue, through of comparison between numerical and experimental data, to the drying of acerolas in the three cases studied. The residue variation to the case 1 estimated by Levenberg-Marquardt method was -1.021 to 1.386, while for the Differential Evolution method was -0.9182 to 1.4763. In case 2 the Levenberg-Marquardt method obtains -0.347 to 0.448 to residue variation, while Differential Evolution method gives -0.2056 to 0.5545. To the case residue variations were -0.306 to 0.198 to Levenberg-Marquardt method, and -0.2008 to 0.2975 to Differential Evolution method.

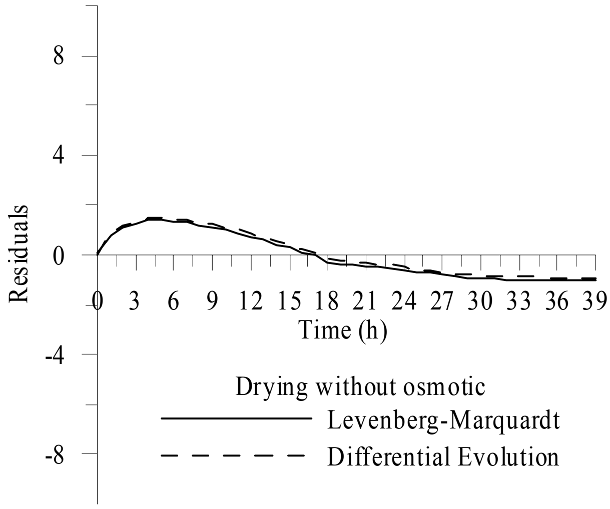

Figure 4. Residues for case 1.

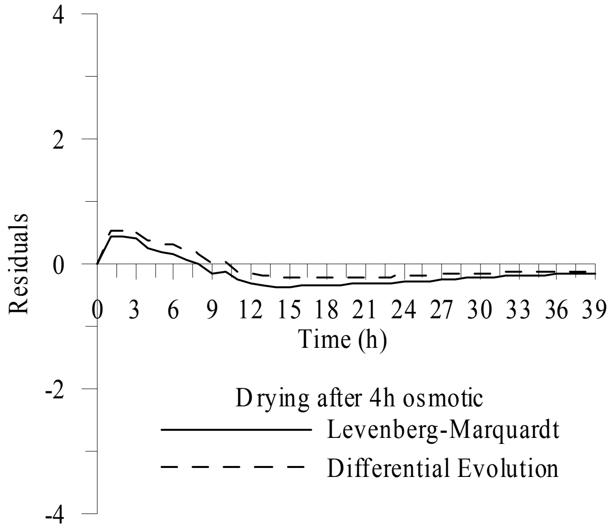

Figure 5. Residues for case 2.

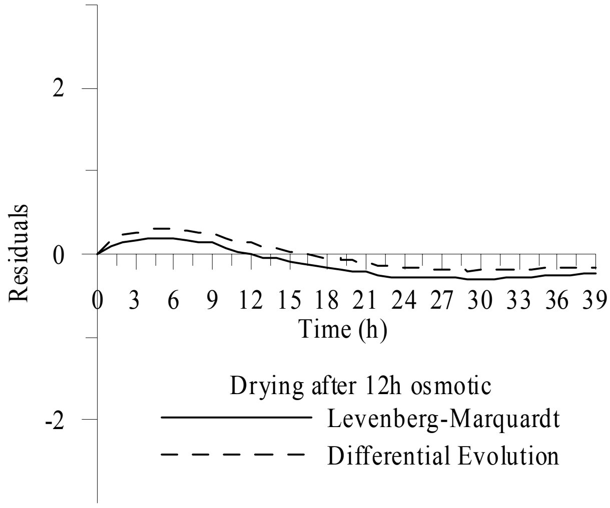

Figure 6. Residues for case 3.

6. CONCLUSION

The diffusional model with constant diffusion coefficient, equilibrium boundary condition and without shrinkage assumption did not adequately represent the West Indian cherry drying process. Nevertheless this model was used in this first work, which has some simplifications. In future works a more complete mathematical model will be adopted. In this study, the effective mass diffusivity of West Indian cherry was estimated by inverse method during drying of West Indian cherry. Inverse methods were not used until now to estimate effective mass diffusivity of West Indian cherry, and in general to estimate thermal physical properties of West Indian cherry. Another real objective of this study was to compare the performance of two optimization methods used to obtain the effective mass diffusivity. Both optimization methods presented good performance, however small discrepancies were observed in cases 1 and 3, than can be explained by hypotheses assumed in the solution of the Fick's 2^{nd} law and also by operational conditions used.

7. ACKNOWLEDGEMENTS

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9. RESPONSIBILITY NOTICE

The authors Mirtes Aparecida da Conceição Silva, Zaqueu Ernesto da Silva e Viviana Cocco Mariani are the only responsible for the printed material included in this paper.