PERIODICA POLYTECHNICA SER. CHEM. ENG. VOL. 46, NO. 1–2, PP. 83–92 (2002)

MODELLING OF SUGAR TRANSFER DURING OSMOTIC DEHYDRATION OF CARROTS

Anikó MATUSEK and Péter MERÉSZ

Department of Biochemistry and Food Technology
Budapest University of Technology and Economics
H–1521 Budapest, Hungary

Received: March 30, 2003

Abstract

Mass transfer was quantitatively investigated during osmotic dehydration of carrot slices and layers. The distribution of water and sugar was determined in longitudinal and radial directions of root samples. Using the solution of Fick's I. and II. laws the effective diffusion coefficients of sugar depending on geometric parameters in radial layers and longitudinal slices were estimated according to cylindrical and rectangular coordinate systems. The observed dependency of $D_{r,s}$ and $D_{l,s}$ was found to be in line with the absence of alteration in longitudinal and the existing difference in radial directions of the tissue structure in carrot root.

Keywords: distribution of diffusion coefficients, sugar uptake, osmotic dehydration.

1. Introduction

Osmotic dehydration is a water removal technique, which is applied to horticultural products, such as fruits and vegetables, to reduce the water content, while increasing soluble solid content [12]. The semi-final product obtained is not stable from the point of view of conservation. The chemical composition and organoleptic characteristics of osmodehydrated fruits and vegetables result in high quality products that can undergo subsequent freezing, dehydrofreezing, air drying or vacuum drying. Using the osmotic dehydration combined with subsequent drying products with good organoleptic quality can be obtained. Energy savings are obtained in the complementary heated air drying.

The raw material is placed into concentrated solutions of soluble solids having higher osmotic pressure and lower water activity. Simultaneous water and solute diffusion processes are caused by the water and solute activity gradients across the cell membrane, the cell wall and the surface of the tissue. The complex cellular structure of food acts as a semipermeable surface. Since these compartments are only partially selective, there is always some solute diffusion into the food. The water transfer is generally accompanied by natural substances (vitamins, flavours, fruit acids, pigments, saccharides, minerals). As a consequence of this exchange, the product loses weight and shrinks. Osmotic dehydration as a pretreatment to many processes improves nutritional, sensorial and functional properties of food without changing its integrity. It is effective even at ambient temperature, so heat damage to texture, colour and flavour of food is minimized [21].

The influence of the main process variables (concentration and composition of the osmotic solution, temperature, immersion time, pre-treatments, agitation, nature of food and its geometry, and solution/sample ratio) on the mass transfer mechanism and product quality have been studied extensively. In spite of the numerous studies it is difficult to establish general rules about the variables that affect osmotic dehydration. It was reported that an increase in concentration and temperature of the osmotic solution increased the rate of mass transfer up to a certain extent, above which undesirable changes in flavour, colour and the texture of the product were observed.

In the last few years, numerous studies have been carried out to better understand the internal mass transfer occurring during osmotic dehydration of foods and to model the mechanism of the process.

The purpose of our project is to study the effect of temperature and concentration of osmotic solution on the extent of osmotic dehydration of carrot, to investigate the mechanism of simultaneous mass transfer during osmotic dehydration, to determine the differences among the apparent diffusion coefficients of sugar in the subsequent layers in the carrot roots.

2. Mathematical Modelling

Two resistances oppose mass transfer during osmotic dehydration of horticultural products, an internal and an external. The fluid dynamics of the solid–fluid interface governs the external resistance, whereas the internal one is influenced by the cell tissue structure and the interaction between the different mass fluxes. Under the usual treatment conditions the external resistance is negligible compared to the internal one [24].

A number of investigators used Fick's unsteady state law of diffusion to estimate the water or solute diffusivity, simulating the experiments with boundary conditions to overcome the assumptions involved in Fick's law [5, 9, 20, 11] . Assumptions include constant concentrations of external solution and negligible surface resistance compared with the internal diffusion resistance [13]. The assumption of constant solution concentration can be satisfied by maintaining a high solution to food ratio. This assumption can be satisfied in laboratory scale. But, many problems can be faced when high volumes of concentrated solutions are to be circulated through the equipment in an industrial application. The solution/food ratio of 4 to 6 is optimum for the optimum osmotic effect [14]. The assumption of negligible external resistance cannot always be satisfied at high viscosity, at low temperature and high solute concentration. It was shown that the external resistance cannot be negligible for osmotic dehydration carried out at different agitation conditions [18].

Solution of Fick's Law of diffusion for short contact time was used by several authors [8, 15, 3]. In this model, the overall mass transfer coefficient was used instead of the diffusion coefficient and it was estimated from the slope of concentration vs square root of time. However, this model is limited in the information to be derived from and not valid for long contact time [19].

While solute transfer is assumed to be of diffusion type, the fact that water loss is greater than solid gain is attributed to an osmotic transport phenomenon across the semipermeable cellular membranes. Thus, the process is termed 'osmotic dehydration', though perhaps the most appropriate term would be 'dehydration driven by concentration differences' or 'dehydration and impregnation by immersion' (proposed by RAOULT-WACK) [22]. According to some authors, solute penetration is confined to extracellular spaces [8, 4, 16] proved by microscope observation that sucrose passes through the cell wall and accumulates between the cell wall and the cellular membrane [10, 23].

As a lot of facts seem to indicate that several transfer mechanisms coexist during the osmotic dehydration of horticultural products, the development of a mathematical model that is able to include all these mechanisms is very difficult. There are numerous mathematical models that are capable of representing the experimental data but their use is limited to certain cases and they do not take into account the mechanism on which the results depend.

A two-parameter kinetic model is proposed, based on mass balance, which was used to estimate mass transfer coefficients and the final equilibrium point [1]. This model was able to predict water loss and solid gain at equilibrium conditions using the experimental data obtained during a relatively short period of time. A model was developed incorporating cell membrane characteristics for the simulation of water and solute fluxes in complex cellular tissue [25, 26]. Later, a model was developed based on thermodynamic description of the forces in the osmotic process [17]However, this model depends on a large number of biophysical properties, such as elastic modulus of the cell wall, cell wall void fraction, cell wall tortuosity and membrane permeabilities, which are very difficult to measure or to find in the literature for food materials.

Fick's Law for unsteady state diffusion, as a former model, can be described as the molecular mass transport equation:

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

where C is the concentration of the diffusing component, D is the diffusion coefficient, t is the time coordinate, x is the space coordinate, the diffusion path. The double mass transfer is reduced this way to only one transport process, either of water or solute.

The different analytical solutions of the Fick Law found by CRANK [6] for several geometries and boundary conditions relate the amount of mass transferred, in terms of a series of exponential functions:

$$\frac{M}{M_\infty} = 1 - \sum_{n=\infty}^{m=0} f_1 \exp\left(-f_2\frac{D\cdot t}{t^2}\right), \tag{2}$$

where M is the mass of diffusing substance that traversed the membrane and M_∞ is the same for equilibrium conditions. Functions f_1 and f_2 depend on n and on the mass ratio of solution to solid, l is the characteristic solid dimension.

There is a series of assumptions used in the models, according to the different circumstances, sample geometries, etc. as follows:

- Slices are infinite slabs of width = $x \cdot r$;
- The syrup to fruit ratio is sufficiently high, so that the syrup concentration can be considered constant;
- Initial water and soluble solids concentrations are uniform;
- Apparent diffusion coefficient is constant ($D \neq f(C)$);
- Simultaneous counter-current flows. The diffusion of the water and the sugar are only considered. Other mass transfers are neglected;
- External resistance to mass transfer is neglected;
- The surface resistance is significant and the fluxes at the surface in a fixed frame are proportional to the concentration difference;
- There is no resistance at the surface and a sugar solution film is assumed to exist as a boundary layer;
- Shrinkage occurs in the same proportion in all directions, or
- Shrinkage is neglected during osmotic dehydration;
- The process is isothermal.

Also the dimension of M used varies from one author to another. On the other hand the authors have obtained the apparent diffusion coefficient to reach the best fit with the experimental data. CONWAY [5] found the value of D of water ranging from 15^*10^{-9} to 60^*10^{-9} m^2/s, BERISTAIN [2] obtained the value of D ranging from 0.6^*10^{-9} to 2.5^*10^{-9} m^2/s.

Knowledge of water activity during osmotic dehydration is also a useful parameter because of its relationship with microbial growth. From average sugar concentrations predicted by the model, water activity values can be calculated using FAVETTO and CHIRIFE's correlation [7]: $a_w = 1 - K \times M$, where a_w is the water activity, K is constant, which varies according to the solute (0.0196 mol for glucose and 0.0248 mol for sucrose) and M is sugar concentration in the fruit expressed as molality (moles sugar/kg water).

3. Materials and Methods

3.1. Raw Material Preparation

The carrots (cv.Nanti) and the sucrose (commercial) were purchased from a local supermarket. The carrots were classified by size and shape to have uniformity of raw material. The carrots after peeling were cut into circular pieces of diameter 40 mm and height 40 or 70 mm depending on the direction of the diffusion. After a certain time of heat treatment at 90 °C in 15 °B water and drying, a plastic tube with 4 mm diameter was placed in the middle of the sample to establish the conditions of the limited diffusion in the direction of radius.

3.2. Experimental Design

The samples were placed in osmotic solution with 30 °B, the rate of the solution to the sample mass was 3:1, and it was agitated at 25 °C. Sampling was after 1, 2, 3, 4, 6, 10 and 20 hs of immersion, samples were taken out, quickly rinsed and gently blotted dry with tissue paper to remove adhering osmotic solution and then the samples were cut into 2 mm slices for measuring the longitudinal diffusion (7 *slices* per sample), or peeled to 2 mm thick layers for measuring the radial diffusion (7 *layers* per sample). The half of the samples were dried at 70 °C till a constant weight loss was obtained. The other half of samples were prepared for sucrose determination based on the determination of optical activity. In each of the experiments fresh sucrose syrup was used. All the experiments were done in triplicate and average values were taken for calculations.

3.3. Calculations

In the case of longitudinal diffusion the equation derived from Fick Law II. in a rectangular coordinate system was used to determine the diffusion coefficient: $D_{l,s}$.

$$S = -D_{l,s} \cdot \left(\frac{\pi^2}{4x_0^2}\right), \tag{3}$$

where S is the slope of the function $\left[\ln\left(\frac{c_\infty - c}{c_\infty}\right) = f(t)\right]$ fitted to the observed data, $D_{l,s}$ is the longitudinal diffusion coefficient of the solid [m^2/s], x_0 is the geometric parameter, the diffusion distance [m].

In the case of radial diffusion the solution of Fick Law II. in a cylindrical coordinate system was used to determine the diffusion coefficient: $D_{c,s}$.

$$S = -D_{r,s} \frac{I_\infty^2}{(R - R_b)^2}, \tag{4}$$

where S is the slope of the function $\left[\ln\left(\frac{c_\infty - c}{c_\infty}\right) = f(t)\right]$ fitted to the observed data, $D_{r,s}$ is the radial diffusion coefficient of the solid [m^2/s], I_∞ is the abscissa value at first zero ordinate value of the zero order Bessel function ($I_\infty = 2.4048$), R is the radius of the carrot sample [m], R_b is the radius of the plastic tube in the centre of the carrot [m], $(R - R_b)$ difference is the geometric parameter, the diffusion distance [m].

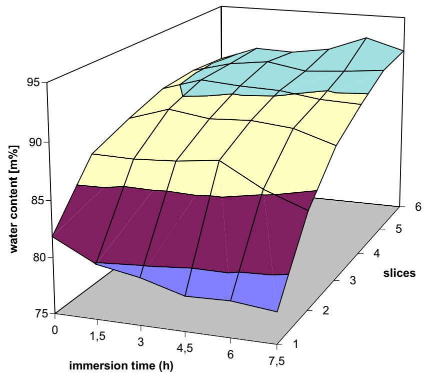

Fig. 1. Changes in radial distribution of water in carrot root

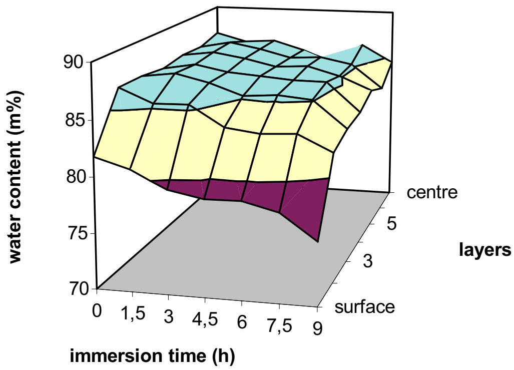

Fig. 2. Changes in longitudinal distribution of water in carrot root

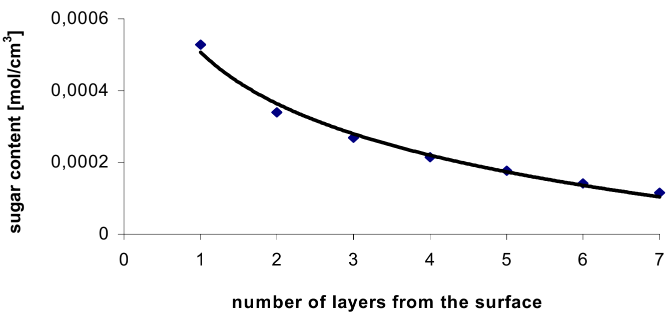

Fig. 3. Radial distribution of the sugar in the carrot layers

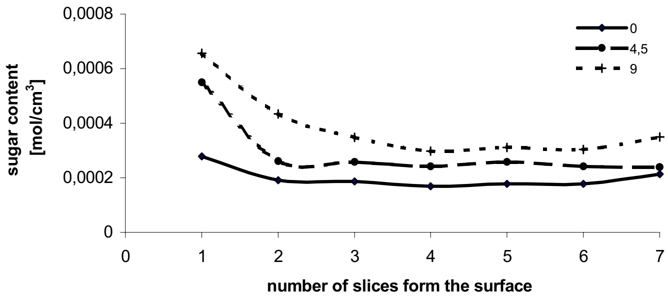

Fig. 4. Changes in longitudinal distribution of the sugar in the carrot slices

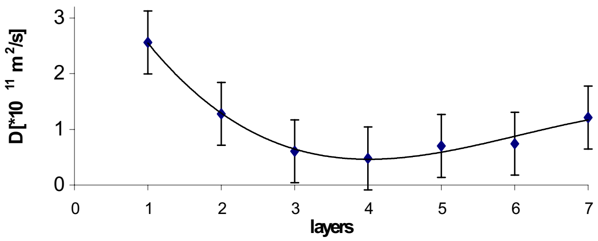

Fig. 5. Radial distribution of diffusion coefficients in the carrot layers

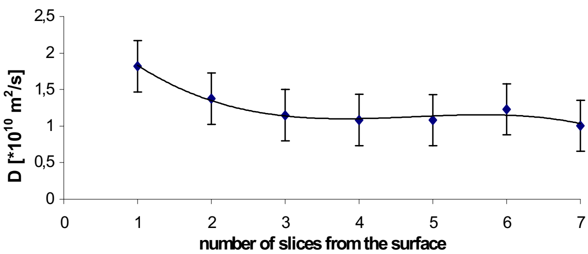

Fig. 6. Longitudinal distribution of the effective diffusion coefficients in the carrot slices

4. Results and Discussion

4.1. The Water Distribution

The average water content of the carrot samples was 78–92 m%, with rel.standard deviation 1.97%. The distribution of water content in radial direction is found as seen layer by layer in *Fig. 1*. The lowest value for water content was found in the outer layer and the highest in the centre of the carrot. The distribution of water is characterized by logarithmic concentration gradient during the immersion time.

The water concentration of the layers decreases as *Fig. 1* shows.

Similar distribution is observed in the longitudinal direction (*Fig. 2*), but the significance of the distribution has lower possibility. Also the decrease and the alteration in the longitudinal water distribution is well seen. The average water content of the carrot samples was 78–88 m%, with relative standard deviation 0.93%.

4.2. The Sugar Distribution

The longitudinal sugar distribution is opposite to that of the water according to the values in the slices in *Fig. 3*. The sugar concentration changes in the range 0.1 – 0.58 mmol/cm^3. The standard deviation of the sugar determination is 0.025 mmol/cm^3. The radial sugar distribution is shown in *Fig. 4*, having constant concentration from the outer layer (number 1) to the centre (number 7). The sugar content increases during the immersion, and the distribution changes as *Fig.4* shows. The highest rate of the sugar gain is measured in the outer layer. The numbers 0, 4.5 and 9 mean the immersion time in hours.

4.3. The Distribution of Diffusion Coefficients of the Sugar

The sugar diffusion coefficients (D_s) were calculated by the method given in 3.3. The S value on the left side of the *Eqs.* (3) and (4) is calculated from the logarithmic presentation of the data. Both the longitudinal ($D_{l,s}$) and radial ($D_{r,s}$) values were calculated slices by slices and layers by layers, respectively. The calculated effective diffusion coefficients for sugar obtained for different directions and distances from the surface are given in *Table 1*. The distribution of $D_{r,s}$ is presented in *Fig. 5* and the distribution of $D_{l,s}$ is in *Fig. 6*.

$D_{r,s}$ values are larger at the surface of the carrot and at outer layers, that reflects the differences in the root tissue structure, or the decreasing permeability of the layers from the outer layers to the centre. The D values are not different in the inner 3–4 layers of the samples. The hypothesis is accepted at 80% significance level. The $D_{l,s}$ values are not different at this probability according to the homogeneous tissue structure in the longitudinal direction. The average values of the samples are $0.5^{*10} - 3^{*}10^{10}\text{m}^2/\text{s}$, that is similar as published by other researchers.

5. Conclusions

The water and the sugar shows a definite and unexpected geometrical distribution in the carrot root either in longitudinal or in radial direction. Fickian unsteady state diffusion was found to be most appropriate for the osmotic dehydration process of the present case. The observed dependency of $D_{r,s}$ and $D_{l,s}$ was found to be in line with the absence of alteration and existing difference in the tissue structure

Table 1. Effective diffusion coefficients of sugar in carrot

Number of layer or slice	$D_{r,s}$ [$*10^{11}$ m^2/s]	$D_{r,s}$ [$*10^{11}$ m^2/s]	$D_{l,s}$ [$*10^{11}$ m^2/s]	$D_{l,s}$ [$*10^{11}$ m^2/s]
1	2.562	2.108	2.264	1.818
2	1.279	0.127	0.887	1.374
3	0.607	0.291	2.876	1.149
4	0.479	0.413	1.310	1.084
5	0.699	0.355	1.818	1.080
6	0.744	0.955	0.834	1.230
7	1.211	1.138	1.282	1.006

of the carrot root. The diffusion coefficients of sugar estimated and the method of determination are expected to be useful in planning the osmotic dehydration process conditions and in the scale-up.

Acknowledgements

The authors are grateful to their own families for constant encouragement and to Mr. János Bódi, assistant professor, Dept. of Physical Chemistry at Budapest University of Technology and Economics.

References

[1] AZUARA, E. – CORTES,R. – GARCIA, H. S. – BERISTAIN, C. I., Kinetic Model for Osmotic Dehydration and its Relationship with Fick's Second Law, *International Journal of Food Science and Technology,* **27** (1992), pp. 239–242.

[2] BERISTAIN, C. I. – AZUARA, E. – CORTES, R. – GARCIA, H. S., Mass Transfer During Osmotic Dehydration of Pineapple Rings, *International Journal of Food Science and Technology*, **25** (1990), pp. 576–582.

[3] BISWAL, R. N. – BOZORGMEHR, K. – TOMPKINS, F. D. – LIU, X., Osmotic Concentration of Green Peas prior to Freezing, *Journal of Food Science,* **56** (1983), pp. 1008–1012.

[4] BOLIN, H. R. – HUXSOLL, C. C. – JACKSON, R. – NG, K. C., Effect of Osmosis Agents and Concentration on Fruit Quality, *Journal of Food Science*, **48** (1983), pp. 202–205.

[5] CONWAY, J. M. – CASTAIGNE, F. – PICARD, G. – VOXAN, X., Mass Transfer Considerations in the Osmotic Dehydration of Apples, *Canadian Institute of Food Science and Technology,* **16** (1983), pp. 25–29.

[6] CRANK, J., *The Mathematics of Diffusion*, Clarendon Press, Oxford, UK, 2nd Edn., 1979.

[7] FAVETTO, G. J. – CHIRIFE, J., Simplified Method for the Prediction of Water Activity in Binary Aqueous Solutions, *Journal of Food Technology,* **20** (1985), pp. 631–636.

[8] HAWKES, J. – FLINK, J. M., Osmotic Concentration of Fruit Slices prior to Freeze Dehydration, *Journal of Food Processing and Preservation,* **2** (1978), pp. 265–284.

[9] HOUGH, G. – CHIRIFE, J. – MARINI, C. A., A Simple Model for Osmotic Dehydration of Apples, *Lebensmittel Wissenschaft und Technologie*, **26** (1993), pp. 151–156.

[10] ISSE, M. G. – SCHUBERT, H., Osmotic Dehydration of Mango and Syrup, 4^{th} *World Congress of Chemical Engineering*, Karlsruhe, Germany, 1991.
[11] KAYMAK-ERTEKIN, F. – CAKALOZ, T., Osmotic Dehydration of Peas: I. Influence of Process Variables on Mass Transfer, *Journal of Food Processing and Preservation*, **20** (1996), pp. 87–104.
[12] KAYMAK-ERTEKIN, F. – SULTANOGLU, M., Modelling of Mass Transfer During Osmotic Dehydration of Apples, *Journal of Food Engineering*, **46** (2000), pp. 243–250.
[13] LAZARIDES, H. N. – NICKOLAIDIS, A. – KATSANIDIS, E., Sorption Changes Induced by Osmotic Pre-Concentration of Apple Slices in Different Osmotic Media, *Journal of Food Science*, **60** (1995), pp. 348–350.
[14] LENART, A. – FLINK, J. M., Osmotic Concentration of Potato: I. Criteria for End Point of the Osmotic Process, *Journal of Food Technology*, **19** (1984) pp. 45–63.
[15] MAGEE, T. R. A. – HASSABALLAH, A. A. – MURPHY, W. R., Internal Mass Transfer during Osmotic Dehydration of Apple Slices in Sugar Solutions, *International Journal of Food Science and Technology*, **1** (1983), pp. 177–178.
[16] MARCOTTE, M., Mass Transfer Phenomena in Osmotic Processes, Experimental Measurements and Theoretical Considerations, Ph.D. thesis, Univ. of Alberta 1988.
[17] MARCOTTE, M. – TOUPIN, C. J. – LE MAGUER, M., Mass Transfer in Cellular Tissues. Part I: The Mathematical Model, *Journal of Food Engineering*, **13** (1991), pp. 199–220.
[18] MAVROUDIS, N. E. – GEKAS, V. – SJÖHOLM, I., Osmotic Dehydration of Apples – Effects of Agitation and Raw Material Characteristics, *Journal of Food Engineering*, **35** (1998), pp. 191–209.
[19] PARJOKO, M. A. – RAHMAN, M. S. – BUCKLE, K. A. – PERERA, C. O., Osmotic Dehydration Kinetics of Pineapple Wedges Using Palm Sugar, *Lebensmittel Wissenschaft und Technologie*, **29** (1996), pp. 452–459.
[20] RASTOGI, N. K. – RAGHAVARAO, K. S. M. S., Effect of Temperature and Concentration on Osmotic Dehydration of Coconut, *Lebensmittel Wissenschaft und Technologie*, **27** (1994), pp. 564–567.
[21] RASTOGI, N. K. – RAGHAVARAO, K. S. M. S., Water and Solute Diffusion Coefficients of Carrot as a Function of Temperature and Concentration during Osmotic Dehydration, *Journal of Food Engineering*, **34** (1997), pp. 429–440.
[22] RAOULT-WACK, A. L. – GUILBERT, S., La déshydration osmotique ou procédé de déshydration-imprégnation par immersion dans des solutions concentrées, *Les cahiers de l'ENS, BANA 'L'eau dans des procédés de transformation et de conservation des aliments'*, **7** (1990), pp. 171–192.
[23] SAUREL, R., Deshydration-impregnation par immersion en solutions ternaires: Etude des transports d'eau et de solutes sur gel et produits d'origine animale, *Ph.D. thesis, Université de Montpellier II.*, 1995.
[24] SPIAZZI, E. A. – MASCHERONI, R. H., Mass Transfer Model for Osmotic Dehydration of Fruits and Vegetables – I. Development of the Simulation Model, *Journal of Food Engineering*, **34** (1997), pp. 387–410.
[25] TOUPIN, C. J. – LEMAGUER, M., Osmotically Induced Mass Transfer in Plant Storage Tissues. A Mathematical Model – Part 2, *Journal of Food Engineering*, **10** (1989a), pp. 97–121.
[26] TOUPIN, C. J. – MARCOTTE, M. – LEMAGUER, M., Osmotically Induced Mass Transfer in Plant Storage Tissues. A Mathematical Model – Part 1, *Journal of Food Engineering*, **10** (1989b), pp. 13–38.