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Some Problems Related to Mathematical Modelling of Mass Transfer Exemplified of Convection Drying of Biological Materials

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1. Introduction

Drying is a widely used industrial process, consuming 7-15% of total industrial energy production in the industrialized world (Dincer & Dost, 1996). Drying is one of the most common and the oldest ways of biological materials preservation such as vegetables and fruits. The main objective in drying biological materials is the removal of water in the solids up to a certain level, at which microbial spoilage and deterioration chemical reactions are greatly minimalized.

The most important aspect of drying technology is the mathematical modelling of the drying processes and the equipment. Its purpose is to allow design engineers to choose the most suitable operating conditions and then size the drying equipment and drying chamber accordingly to meet desired operating conditions. Full-scale experimentation for different products and systems configurations is sometimes costly or even not possible (Sacilik et al., 2006).

Convection drying of biological products is a complex process that involves heat and mass transfer phenomena between the airflow and the product. Mathematical modelling of the drying process of vegetables, fruits and grass is especially difficult because of high initial moisture content (80-95% w.b.) and occurrence of shrinkage during drying.

The course and time of drying process depend on drying conditions, on temperature and moisture profiles developed during the drying process, and above all, on moisture movement in the material. Moisture movement is governed by the properties, form and size of the product and the type of moisture bond in the material (Sander et al., 2003). The major factors affecting the moisture transport during solids drying can be classified as:

i. external factors: these are the factors related to the properties of the surrounding air such as temperature, pressure, humidity, velocity and area of the exposed surface,
ii. internal factors: these are the parameters related to the properties of the material such as moisture diffusivity, moisture transfer coefficient, water activity, structure and composition, etc. (Dincer & Hussain, 2004).

The development of mathematical models to describe the drying process has been the topic of many research studies for several decades (Sander et al., 2003). Presently, more and more sophisticated drying models are becoming available, but a major question that still remains is the accuracy of predictions of drying processes using mathematical models. It is highly dependent on the completeness of the mathematical model and the relationships used to describe heat and mass transfer phenomena of dried products. However, professional literature provides insufficient information on the mathematical modelling of mass transfer during drying of biological materials with a high initial moisture content. Therefore, the aim of the present chapter was to discuss in detail the main problems related to description of the drying process of such materials using differential transport equations.

2. Differential transport equations for drying

According to the theory of convection drying of bodies with sufficiently high initial moisture content, the process of convection drying should proceed in the first period of drying until the critical moisture content is reached. The factors that influence the drying process in the first period of drying are the conditions of external heat and mass exchange in the system: drying product and surrounding air movement. For the case when moisture content of the body is less than its critical moisture content, the second period of drying begins. The process is decided by internal conditions of mass exchange and inner diffusion of water because the internal resistance to water transfer in the body is greater than the external resistance to water transfer from the body (Pabis, 1999; Pabis & Jaros, 2002).

2.1 The first drying period

The course of the drying process at the first drying period is decided by external conditions of mass transfer. In this period the rate of drying is determined by the following equation (Pabis, 1999):

$$\frac{dM}{dt} = -\frac{hA_0}{W_s L}\left(T_a - T_A\right) = -k = \text{const} \tag{1}$$

Eq. (1) resulted from the assumption that all heat delivered to the solid being dried at the first drying period is used for vaporizing water. The solution of Eq. (1) with the assumption valid for the first period of drying

$$T_A = T_{wb} \tag{2}$$

with initial condition $M(t=0)=M_0$ and with assumption that all parameters on the right side of Eq. (1) are constant is the linear model

$$M\left(t\right) = -kt + M_0 \tag{3}$$

The linear model means the acceptance of the assumption that the shrinkage can be neglected. Biological materials with a high initial moisture content undergo, however, shrinkage and deformation during hot-air drying. When water is removed from such a material, a pressure unbalance is produced between the inner of the material and the external pressure, generating contracting stresses that lead to material shrinkage or collapse, changes in shape and occasionally cracking of the product. Mayor and Sereno (2004) gave a detailed physical description of the shrinkage mechanism and presented a classification of the different models proposed to describe this behaviour in materials with high initial moisture content undergoing drying. The models were classified in two main groups: empirical and fundamental models. Empirical models are convenient and easy to use and therefore they are mainly applied to drying models. Acceptance of the assumption that the surface of dried body changes because of the shrinkage means that A_0 in Eq. (1) should be replaced by A. The solution of such changed Eq. (1) with assumption (2), initial condition $M(t=0)=M_0$, equation

$$\frac{A}{A_0}=\left(\frac{V}{V_0}\right)^{2/3} \tag{4}$$

and with the use of shrinkage model (Pabis, 1999)

$$\frac{V}{V_0}=(1-b)\frac{M}{M_0}+b \tag{5}$$

is the model of the first drying period which takes into account drying shrinkage

$$M(t)=M_0\left[\frac{1}{1-b}\left(1-\frac{1-b}{3M_0}kt\right)^3-\frac{b}{1-b}\right] \tag{6}$$

$$b=\frac{0.85}{1+M_0} \tag{7}$$

Replacing shrinkage model described with Eq. (5) by model proposed by Karathanos (1993)

$$\frac{V}{V_0}=\left(\frac{M}{M_0}\right)^n \tag{8}$$

gives the following model of the first drying period

$$M(t)=\left[\frac{(2n-3)kt+3M_0}{3M_0^{2n/3}}\right]^{3/(3-2n)} \tag{9}$$

Eq. (4) means that the constancy of the body shape during drying is taken into consideration. However, such a situation not always occur in the case of the drying of biological materials with a high initial moisture content and thus the dependence Eq. (4) should also, for this reason, be regarded as approximate. In this connection, the accuracy of calculation of A can be improved by introducing into Eq. (4) an empirical coefficient $n_1 \geq 1$, which changes Eq. (4) to (Pabis, 1999)

$$\frac{A}{A_0}=\left(\frac{V}{V_0}\right)^{2/3n_1} \tag{10}$$

and Eq. (6) (after substitution of $3n_1/(3n_1-2)=N$) to

$$M(t)=M_0\left[\frac{1}{1-b}\left(1-\frac{1-b}{NM_0}kt\right)^N-\frac{b}{1-b}\right] \tag{11}$$

The parameter k, which occurs in the models of the first drying period, determines the initial drying rate.

2.2 The second period of drying

It can be accepted that the water movement inside the dried solid is only a diffusion movement in the convection drying process of biological materials with a high initial moisture content. Therefore the equation applied to the description of the second drying period of biological materials takes the following form (Luikov, 1970):

$$\frac{\partial M}{\partial t} = D\nabla^2 M \tag{12}$$

Certain simplifications were made in this equation. It was assumed that shape and volume of dried particle do not change, and that water diffusion coefficient is constant. (The discussion how to take into account shrinkage and changeability of the water diffusion coefficient will be conducted in Section 2.3).

The following initial and boundary conditions can be adopted:

i. the initial condition: the same moisture content at any point of dried material at the beginning of the second drying period (material before drying is cut into small pieces and therefore this assumption can be accepted)

$$M\big|_{t=0} = M_c \tag{13}$$

ii. one of the three boundary conditions can be taken:
   - the boundary condition of the first kind means that the external resistance to mass transfer is negligible (i.e., the surface of the solid is at equilibrium with the surrounding air for the time considered) and therefore the moisture content on the solid surface is equal to the equilibrium moisture content

$$M\big|_A = M_e \tag{14}$$

   - the boundary condition of the second kind means that the mass (water) flux from the surface of the solid is known for the time considered

$$\dot{W}\big|_A = f_A(t) \tag{15}$$

   - the boundary condition of the third kind means that the mass (water) flux from the surface of the solid is expressed in terms of moisture content difference between the surface and the equilibrium moisture content

$$-D\frac{\partial M}{\partial n}\bigg|_A = h_m\left(M\big|_A - M_e\right) \tag{16}$$

For the mass transfer at the surface of the biological materials with a high initial moisture content being dried the first kind and the third kind boundary conditions are mostly used in mathematical models of the drying process (Markowski, 1997).

Biological materials before drying are cut into small pieces, mostly slices or cubes. Therefore Eq. (12) applied to the description of the second drying period of biological materials takes the following form (Luikov, 1970):

for an infinite plane (slices):

$$\frac{\partial M}{\partial t} = D \frac{\partial^2 M}{\partial x^2} \qquad (17)$$
$$\left(t > 0; -R_c < x < +R_c\right)$$

for a finite cylinder (slices):

$$\frac{\partial M}{\partial t} = D\left(\frac{\partial^2 M}{\partial r^2} + \frac{1}{r}\frac{\partial M}{\partial r} + \frac{\partial^2 M}{\partial z^2}\right) \qquad (18)$$
$$\left(t > 0; 0 < r < R_c; -h < z < +h\right)$$

for cubes:

$$\frac{\partial M}{\partial t} = D\left(\frac{\partial^2 M}{\partial x^2} + \frac{\partial^2 M}{\partial y^2} + \frac{\partial^2 M}{\partial z^2}\right) \qquad (19)$$
$$\left(t > 0; -R_1 < x < +R_1; -R_2 < y < +R_2; -R_3 < z < +R_3\right)$$

The initial conditions (Eq. (13)) are following:
for an infinite plane

$$M(x,0) = M_c = \text{const.} \qquad (20)$$

for a finite cylinder

$$M(r,z,0) = M_c = \text{const.} \qquad (21)$$

for cubes

$$M(x,y,z,0) = M_c = \text{const.} \qquad (22)$$

The boundary conditions of the third kind (Eq. (16)) take following form:
for infinite plane

$$\pm D \frac{\partial M(\pm R_c, t)}{\partial x} = h_m\left[M(\pm R_c, t) - M_e\right] \qquad (23)$$

for a finite cylinder

$$-D \frac{\partial M(R_c, z, t)}{\partial r} = h_m\left[M(R_c, z, t) - M_e\right] \qquad (24)$$

$$\frac{\partial M(0,z,t)}{\partial r} = 0,\ M(0,z,t) \neq \infty \qquad (25)$$

$$-D \frac{\partial M(r,h,t)}{\partial z} = h_m\left[M(r,h,t) - M_e\right] \qquad (26)$$

$$\frac{\partial M(r,0,t)}{\partial z} = 0 \tag{27}$$

for cubes

$$\pm D \frac{\partial M(\pm R_1, y, z, t)}{\partial x} = h_m \left[M(\pm R_1, y, z, t) - M_e \right] \tag{28}$$

$$\pm D \frac{\partial M(x, \pm R_2, z, t)}{\partial y} = h_m \left[M(x, \pm R_2, z, t) - M_e \right] \tag{29}$$

$$\pm D \frac{\partial M(x, y, \pm R_3, t)}{\partial z} = h_m \left[M(x, y, \pm R_3, t) - M_e \right] \tag{30}$$

An analytical solution of: (i) Eq. (17) at the initial and boundary conditions given by Eqs. (20) and (23), (ii) Eq. (18) at the initial and boundary conditions given by Eqs. (21) and (24)-(27), and (iii) Eq. (19) at the initial and boundary conditions given by Eqs. (22) and (28)-(30) with respect to mean moisture content as a function of time, take the following form (Luikov, 1970):
for an infinite plane

$$\frac{M(t) - M_e}{M_c - M_e} = \sum_{i=1}^{\infty} B_i \exp\left(-\mu_i^2 \frac{D}{R_c^2} t \right) \tag{31}$$

where

$$B_i = \frac{2Bi^2}{\mu_i^2 \left(Bi^2 + Bi + \mu_i^2 \right)}; \ \mathrm{ctg}\ \mu_i = \frac{1}{Bi} \mu_i$$

for a finite cylinder

$$\frac{M(t) - M_e}{M_c - M_e} = \sum_{i=1}^{\infty} \sum_{j=1}^{\infty} B_{i,1} B_{j,2} \exp\left[-\left(\frac{\mu_{i,1}^2}{R_c^2} + \frac{\mu_{j,2}^2}{h^2} \right) Dt \right] \tag{32}$$

where

$$B_{i,1} = \frac{2Bi^2}{\mu_{i,1}^2 \left(Bi^2 + Bi + \mu_{i,1}^2 \right)} \quad B_{j,2} = \frac{2Bi^2}{\mu_{j,2}^2 \left(Bi^2 + Bi + \mu_{j,2}^2 \right)}$$

$$\mathrm{ctg}\ \mu_{i,1} = \frac{1}{Bi} \mu_{i,1}; \ \mathrm{ctg}\ \mu_{j,2} = \frac{1}{Bi} \mu_{j,2}$$

for cubes

$$\begin{aligned} \frac{M(t) - M_e}{M_c - M_e} &= \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \sum_{k=1}^{\infty} B_{n,1} B_{m,2} B_{k,3} \times \\ &\times \exp\left[-\left(\frac{\mu_{n,1}^2}{R_1^2} + \frac{\mu_{m,2}^2}{R_2^2} + \frac{\mu_{k,3}^2}{R_3^2} \right) Dt \right] \end{aligned} \tag{33}$$

where

$$B_{n,1} = \frac{2Bi^2}{\mu_{n,1}^2\left(Bi^2 + Bi + \mu_{n,1}^2\right)} \quad B_{m,2} = \frac{2Bi^2}{\mu_{m,2}^2\left(Bi^2 + Bi + \mu_{m,2}^2\right)}$$

$$B_{k,3} = \frac{2Bi^2}{\mu_{k,3}^2\left(Bi^2 + Bi + \mu_{k,3}^2\right)}$$

$$\operatorname{ctg} \mu_{n,1} = \frac{1}{Bi}\mu_{n,1}; \operatorname{ctg} \mu_{m,2} = \frac{1}{Bi}\mu_{m,2}; \operatorname{ctg} \mu_{k,3} = \frac{1}{Bi}\mu_{k,3}$$

and for a cubic geometry $R_1=R_2=R_3$.
In the literature on drying boundary conditions of the first kind are almost always taken into account (it is easier to obtain the solution using such boundary conditions) and like for boundary conditions of the third kind the infinite series constitute appropriate solutions. Several researches showed that for large drying times an analytical solution of Eq. (12) for sphere at the appropriate initial condition and boundary condition of the first kind with respect to mean moisture content, can be reduced to the first term of infinite series (Jayas et al., 1991). Pabis et al. (1998) found the relationship between the optimum number of terms in infinite series and Fourier number for sphere and stated that the optimum number of terms increases with the decreasing value of Fo. González-Fésler et al., (2008) demonstrated that for values of Fourier number Fo ($Fo=Dt/R_c^2$)≥0.1 the mathematical solution for the finite cylinder drying including only the first term of each infinite series represents 95% of the complete solution, so that terms with n>1 could be neglected. The number of terms in analytical solution of Eq. (12) for an infinite plane (at the appropriate initial condition and boundary condition of the first kind with respect to mean moisture content) necessary for calculating the moisture ratio MR with accuracy $\delta=4\%$ is i=5 for the initial phase of drying (Fo=0), whereas i=20 and i=193 are needed to achieve an accuracy of 1% and 0.1% respectively. The accuracy was defined as $100(M_\infty-M_i)/M_\infty$, where M_∞ and M_i are the exact and truncated solutions, respectively (Efremov et al., 2008).

2.3 Determination of the parameters necessary for using the models of the second drying period

Knowledge of the value of the critical moisture content, equilibrium moisture content, water (moisture) diffusion coefficient, mass transfer coefficient and Biot number is necessary for using model of the second drying period.
The critical moisture content M_c could be determined assuming the continuity of the drying process. The continuity of the process required that when M is equal to M_c, the drying rate in the first and second period be equal, that is when $(dM/dt)_I=(dM/dt)_{II}$ (Jaros & Pabis, 2006). Górnicki & Kaleta (2007a) used the drying rate and the temperature of the dried particle as a criterion of division into the first and the second drying period. The modelling of the second drying period following the first drying period requires introducing into Eqs. (31), (32), and (33) structure of corrected drying time t minus t_c, where t_c is the drying time to the critical moisture content.
Equilibrium moisture content represents the moisture content that the material will attain if dried for an infinite time at a particular relative air humidity and temperature. The relation

between the material moisture content and the relative humidity in equilibrium with the product at the same temperature used to reach the equilibrium is termed the sorption isotherm (the equilibrium moisture content – equilibrium relative humidity relationship). The sorption isotherms of biological and food materials are the sigmoid shape, under the Brunauer classification (Brunauer, 1943) of type II. The existing isotherm equations can be divided into two separate groups: (i) empirical or partly empirical equations using exponential, power or logarithmic functions and (ii) equations with some theoretical basis and/or their combinations (Blahovec, 2004). It turned out that at least seventy seven isotherm equations are available in the literature (van den Berg & Bruin, 1981). The commonly used equations for biological materials are: the Langmuir, Brunauer-Emmett-Teller (BET), Iglesias-Chirife, the modified Henderson, Chen, Chung-Pfost, Halsey, Oswin, and Guggenheim-Anderson-de Boer (GAB) (Rizvi, 1995). The problems related to fitting abilities of the existing isotherm equations for biological materials and selecting the best equations are still under discussion (Castillo et al., 2003; Furmaniak et al., 2007; Kaleta & Górnicki, 2007; Timmermann et al., 2001).

The value of the equilibrium moisture content is relatively small (especially for low air relative humidity) compared to M(t) or M_0 and therefore the dimensionless moisture content (moisture ratio) $MR=[M(t)-M_e]/(M_0-M_e)$ for the whole process of drying could be simplified to $M(t)/M_0$ (Doymaz & Pala, 2002; Zielinska & Markowski, 2007).

In biological materials with a high initial moisture content water can be transported by water diffusion, vapour diffusion, Knudsen diffusion, internal evaporation and condensation effects, capillary flow, and hydrodynamic flow. Often there is a mixture of various transport mechanisms, and the contributions of the different mechanisms to the total transport varies from place to place and changes as drying progresses (Bruin & Luyben, 1980). Therefore the water diffusion coefficient in the model of the second drying period describes the total transport of moisture and is called the effective diffusivity. The values of moisture diffusion coefficient for biological materials reported in the literature lie within the range of 10^{-12} – 10^{-8} m^2s^{-1} (mostly about 10^{-10} m^2s^{-1}) (Doulia et al., 2000; Maroulis et al., 2001). It is known that moisture diffusion coefficient depends strongly on temperature (e.g. Cuningham et al., 2007; Garcia-Pascual et al., 2006; Kaymak-Ertekin, 2002) and often very strongly indeed on the moisture content (e.g. Maroulis et al., 2001; Waananen & Okos, 1996) and on material structure (e.g. Ruiz-Cabrera et al., 1997). Nevertheless, there is no diffusion theory that is sufficiently well formulated and verified. Therefore the most commonly used method for determining the moisture diffusion coefficient in biological materials is by fitting the diffusion-based drying equations to the experimental data in the second drying period. It should be emphasized, however, that moisture diffusion coefficient determined with this method is limited in application to the diffusion-based drying equation from which it was calculated and to the moisture range in which experiments were conducted (Pabis et al., 1998). In the case of shrinkage and changeability of the water diffusion coefficient the coefficient is determined from the diffusion-based drying equation applying the method of inverse problem (Jaros et al., 1992; Górnicki & Kaleta, 2004).

In the literature concerning mathematical modelling of convection drying of biological materials the value of the water diffusion coefficient is mostly considered as a constant. An Arrhenius – type equation is sometimes used to describe the relationship between the diffusion coefficient and temperature of dried material:

$$D = D_0 \exp\left[-E_a R^{-1} \left(T + 273.15 \right)^{-1} \right] \tag{34}$$

Mulet et al. (1989a,b) expressed the water diffusion coefficient by the following empirical formula:

$$D = a \cdot \exp\left[b\left(T + 273.15 \right)^{-1} \right] \tag{35}$$

The water diffusion coefficient as a function of moisture content and dried material temperature was described by Mulet et al. (1989a,b):

$$D = \exp\left[a + b\left(T + 273.15 \right)^{-1} + cM \right] \tag{36}$$

and Parti & Dugmanics (1990):

$$\frac{D}{R^2} = a \cdot \exp\left(\frac{-b}{T + 273.15} + cM \right) \tag{37}$$

Dincer and Dost (1996) developed and verified analytical techniques to characterise the mass transfer during the drying of geometrically (infinite slab, infinite cylinder, sphere) and irregularly (by use of a shape factor) shaped objects. Drying process parameters, namely drying coefficient S and lag factor G:

$$\frac{M(t) - M_e}{M_c - M_e} = G \exp(-St) \tag{38}$$

were introduced based on an analogy between cooling and drying profiles, both of which exhibit an exponential form with time. The moisture diffusivity D was computed using:

$$D = \frac{SR^2}{\mu_1^2} \tag{39}$$

The coefficient μ_1 was determined by evaluating the root of the corresponding characteristic equation (Dincer et al., 2000):
for slab shapes:

$$\mu_1 = -419.24G^4 + 2013G^3 - 3615.8G^2 + 2880.3G - 858.94 \tag{40}$$

for cylindrical shapes:

$$\mu_1 = -3.4775G^4 + 25.285G^3 - 68.43G^2 + 82.468G - 35.638 \tag{41}$$

for spherical shapes:

$$\mu_1 = -8.3256G^4 + 54.842G^3 - 134.01G^2 + 145.83G - 58.124 \tag{42}$$

Babalis & Belessiotis (2004) used the following method of calculation of effective moisture diffusivity. If following assumptions are accepted in Eq. (31):

i. the external mass transfer resistance is negligible, but the internal mass transfer resistance is large ($Bi \to \infty$),
ii. the first term of infinite series is taken into account, successive terms are small enough to be neglected,

its simplified form can be expressed as follows:

$$\frac{M(t)-M_e}{M_c-M_e}=\frac{6}{\pi^2}\exp\left(-\pi^2\frac{Dt}{R^2}\right) \tag{43}$$

Logarithmic simplification of Eq. (43) leads to a linear form:

$$\ln\left[\frac{M(t)-M_e}{M_c-M_e}\right]=\ln\left(\frac{6}{\pi^2}\right)-\left(\pi^2\frac{Dt}{R^2}\right) \tag{44}$$

By plotting the measured data plotted in a logarithmic scale, the effective moisture diffusivity was calculated from the slope of the line k_1 as presented:

$$k_1=\frac{\pi^2 D}{R^2} \tag{45}$$

Local mass (water) flux on the external surface A of the dried solid biological material, can be described with the equation (right side of Eq. (16)):

$$\dot{W}=h_m\left(M|_A-M_e\right) \tag{46}$$

The mass transfer coefficient can be determined by the following equations (Markowski, 1997; Simal et al., 2001; Magge et al., 1983):

$$h_m=-\frac{V}{A_w}\frac{\partial\bar{M}/dt}{M|_A-M_e} \tag{47}$$

$$h_m=\frac{4}{2\pi R}D_{wa} \tag{48}$$

$$h_m=aC^bT^c \tag{49}$$

The mass transfer coefficient can be also calculated from the dimensionless Sherwood number Sh. The Sherwood number can be expressed:

i. for forced convection as a function of the Reynolds number Re and the Schmidt number Sc (Beg, 1975)

$$Sh=a\,Re^b\,Sc^c \tag{50}$$

$$Sh=a+b\,Re^c\,Sc^d \tag{51}$$

$$Sh=Sh_0+a\,Re^b\,Sc^c \tag{52}$$

ii. for natural convection as a function of the Grashof number (mass) Gr_m and the Schmidt number Sc (Sedahmed, 1986; Schultz, 1963):

$$Sh = aGr_m^b Sc^c \tag{53}$$

$$Sh = 2 + aGr_m^b Sc^c \tag{54}$$

iii. for vacuum-microwave drying as a function of the Archimedes number Ar and the Schmidt number Sc (Łapczyńska-Kordon, 2007)

$$Sh = a(Ar \cdot Sc)^b \tag{55}$$

The dimensionless moisture distributions for three shapes of products are given in a simplified form as Eq. (38) and for:
slab shapes:

$$G = \exp\left(\frac{0.2533Bi}{1.3 + Bi}\right) \tag{56}$$

cylindrical shapes:

$$G = \exp\left(\frac{0.5066Bi}{1.7 + Bi}\right) \tag{57}$$

and spherical shapes:

$$G = \exp\left(\frac{0.7599Bi}{2.1 + Bi}\right) \tag{58}$$

Using the experimental drying data taken from literature sources for different geometrical shaped products (e.g. slab, cylinder, sphere, cube, etc.), Dincer & Hussain (2004) obtained the Biot number–lag factor correlation for several kinds of food products subjected to drying as (R^2= 0.9181):

$$Bi = 0.0576G^{26.7} \tag{59}$$

The dimensionless Biot number Bi for moisture transfer can be calculated using its definition as:

$$Bi = \frac{h_m R}{D} \tag{60}$$

2.4 Equation of heat balance of dried biological material heating

Heat supplied to the particles of dried biological material is used to increase the particle temperature and to vaporize water. Material before drying is cut into small pieces (slices, cubes). It turned out from the experiments that the average value of the dried particle temperature did not differ in essential manner from the temperature value of the solid surface at any instant during process (Górnicki & Kaleta, 2002; Pabis et al., 1998). Therefore equation of heat balance of the dried solid heating obtains the following form (Górnicki & Kaleta, 2007b):

$$c(M+1)\frac{dT}{dt} = -\frac{hA}{\rho_s V_s}(T - T_a) + L\frac{dM}{dt} \tag{61}$$

The specific heat of biological materials with a high initial moisture content depends on composition of the material, moisture content and temperature. Typically the specific heat increases with increasing moisture content and temperature and linear correlation between specific heat and moisture content in biological materials is observed mostly. Most of the specific heat models for discussed materials are empirical rather than theoretical. The present state of the empirical data is not precise enough to support more theoretically based models which in some cases are very complicated. Kaleta (1999) presented a classification of the different specific heat models of biological materials with a high initial moisture content.
Shrinkage model (e.g. Eq. (5) or Eq. (8)) and expression (4) or (10) can be used for determination of the surface area of dried solid presented in Eq. (61).
The heat transfer coefficient can be calculated from the dimensionless Nusselt number Nu. The Nusselt number can be expressed:

i. for forced convection as a function of the Reynolds number Re and the Prandtl number Pr

$$\mathrm{Nu} = a\,\mathrm{Re}^{b}\,\mathrm{Pr}^{c} \tag{62}$$

ii. for natural convection as a function of the Grashof number Gr and the Prandtl number Pr

$$\mathrm{Nu} = a\left(\mathrm{Gr}\cdot\mathrm{Pr}\right)^{b} \tag{63}$$

The constants a, b, and c can be found in Holman (1990).
For materials of moisture content above approximately 0.14 d.b. it can be assumed that to overcome the attractive forces between the adsorbed water molecules and the internal surfaces of material the same energy is needed as heat required to change the free water from liquid to vapour (Pabis et al., 1998).
Eq. (61) can be used for temperature modelling of biological materials during the second drying period.
According to the theory of drying the initial temperature of dried material reaches the psychrometric wet-bulb temperature T_{wb} (Eq. (2)) and remains at this level during the first period of drying. Beginning with the second period of drying, the temperature of material continuosly increases (Eq. (61)) and if the drying lasts long enough, the temperature reaches the temperature of the drying air.

3. Discussion of some results of modelling convection drying of parsley root slices

The authors' own results of research are presented in this chapter.
Cleaned parsley roots were used in research. Samples were cut into 3 mm slices and dried under natural convection conditions. The temperature of the drying air was 50°C. The following measurements were replicated four times under laboratory conditions: (i) moisture content changes of the examined samples during drying, (ii) temperature changes of the examined samples during drying, (iii) volume changes of the examined samples during drying. Measurements of the moisture content changes were carried out in a laboratory dryer KCW-100 (PREMED, Marki, Poland). The samples of 100 g mass were dried. Such a mass ensured final maximum relative error of evaluation of sample moisture content not exceeding 1 %. The mass of samples during drying and dry matter of samples

were weighed with the electronic scales WPE-300 (RADWAG, Radom, Poland). The changes of temperature of samples undergoing drying were measured by thermocouples TP3-K-1500 (NiCr-NiAl of 0.2 mm diameter, CZAKI THERMO-PRODUCT, Raszyn, Poland). Absolute error of temperature measurement was 0.1°C and maximum relative error was 0.7 %. Measurements of moisture content changes and the temperature changes were done at the same time. The volume changes of parsley root slices during drying were measured by buoyancy method using petroleum benzine. Maximum relative error was 5 %.

Figure 1 shows drying curve and changes of the temperature during drying of parsley root slices. The drying curve represents empirical formula approximating results of the four measurement repetitions of the moisture content changes in time.

Figure 2 presents the changes of the temperature during drying of parsley root slices and the results of the temperature modelling using Eq. (61).

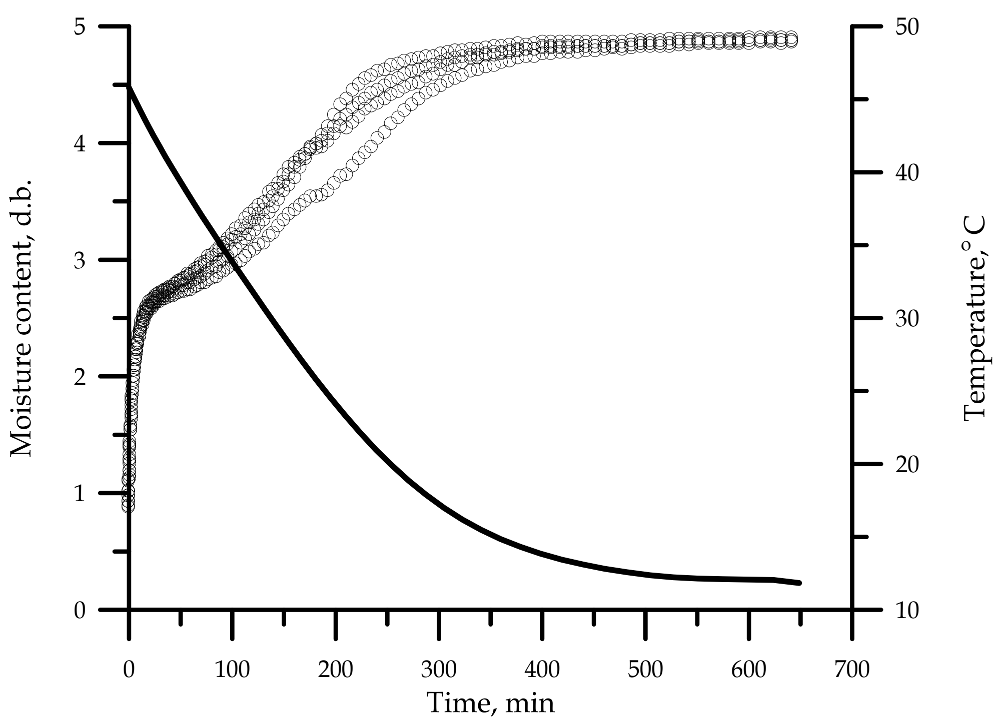

Fig. 1. Moisture content vs. time and temperature vs. time for drying of 3 mm thick parsley root slices at 50°C under natural convection condition: (▬) – empirical formula approximating moisture content changes in time, (○) – temperature

At the beginning of the drying, temperature of slices increases rapidly because of heating of the materials. Then, for some time temperature is almost constant and afterwards slices temperature rises quite rapidly, attaining finally temperature of the drying air. The occurrence of period of almost constant temperature suggests that during drying of parsley root slices there is a period of time during which the conditions of external mass transfer determine course of the process. It can be seen from Fig. 2 that Eq. (61) predicts the temperature of parsley root slices during second period of drying quite well.

The course of drying curve of parsley root slices at the first drying period was described with Eqs. (3), (9), and (11), respectively. Following statistical test methods were used to evaluate statistically the performance of the drying models:

the determination coefficient R^2

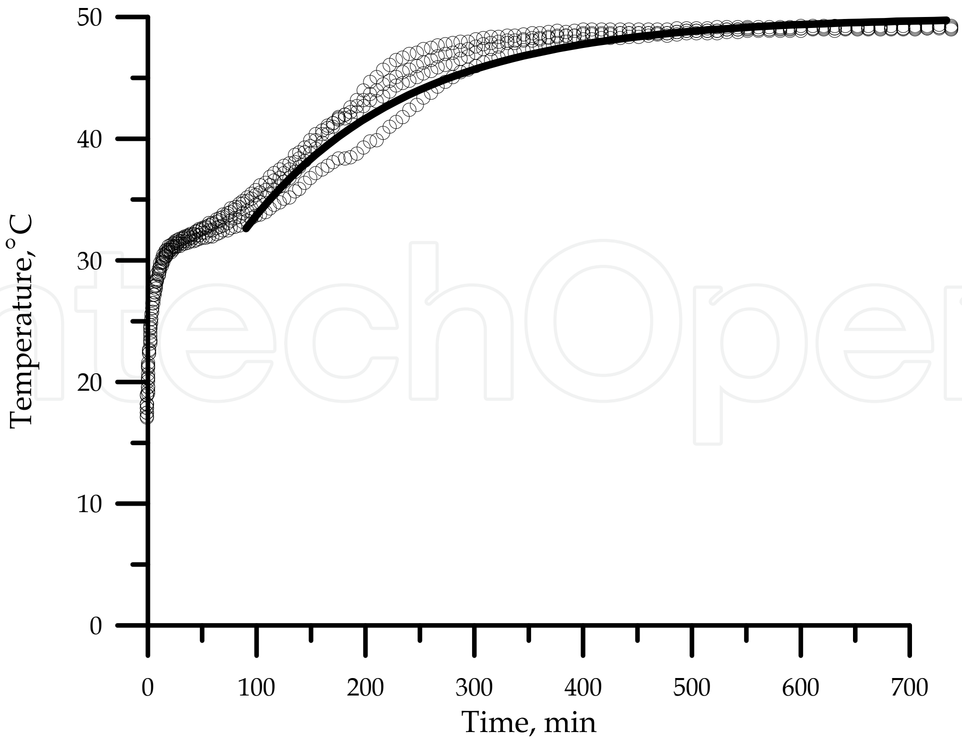

Fig. 2. Changes of the temperature during drying of 3 mm thick parsley root slices at 50°C under natural convection condition: (○) – experimental data, (▬) – Eq. (61)

$$R^2 = \frac{\sum_{i=1}^{N}\left(MR_i - MR_{pre,i}\right)\cdot\sum_{i=1}^{N}\left(MR_i - MR_{exp,i}\right)}{\sqrt{\left[\sum_{i=1}^{N}\left(MR_i - MR_{pre,i}\right)^2\right]\cdot\left[\sum_{i=1}^{N}\left(MR_i - MR_{exp,i}\right)^2\right]}} \tag{64}$$

and the root mean square error RMSE

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N}\left(MR_{pre,i} - MR_{exp,i}\right)^2\right]^{1/2} \tag{65}$$

The higher the value of R^2, and lower the value of RMSE, the better the goodness of the fit. Coefficients of the models of the first drying period and the results of the statistical analyses are given in Table 1.

Model of the first drying period	Coefficients	R^2	RMSE
Eq. (3)	k=0.0164	0.998	0.0224
Eq. (9)	k=0.0164; n=0.7829	0.999	0.0097
Eq. (11)	k=0.0164; b=0.15531; N=2.6	0.999	0.0165

Table 1. Coefficients of the models of the first drying period and the results of the statistical analyses

It was assumed that the models describe drying kinetics correctly when values of the relative error of model (3) do not exceed 1 %, and of models (9) and (11) do not exceed 3 %. A decision was taken to increase the value of the relative error to 3 % due to the nature of the course of the relative error for the models with drying shrinkage. At first, the relative error for these models reached negative value, afterwards it increased reaching zero value and then grew rapidly. As can be seen from the statistical analysis results, high coefficient of determination R^2 and low values of RMSE were found for all models. Therefore, it can be stated that all considered models may be assumed to represent the drying behaviour of parsley root slices in the first drying period.

It turned out that models of the first drying period describe the course of drying curve in different ranges of application. The linear model Eq. (3) describes the process for 80 min but the models of the first drying period which take into account drying shrinkage Eqs. (9) and (11) describe the process for 340 min and 305 min, respectively. Comparison with the course of the slices temperature (Fig. 1) points towards the following conclusions: (i) the linear model describes the drying from the beginning of the process till the end of period of constant temperature, (ii) models with shrinkage describe the process till the moment when slices temperature almost approach to drying air temperature. The analysis of the results obtained indicates that the course of the whole drying curve of parsley root slices could be described satisfactorily by using only the models with drying shrinkage. Such a description can be useful from the practical point of view because the solution of the model with drying shrinkage is easy to obtain.

The course of drying curve of parsley root slices at the second drying period was described with Eq. (31). Biot number Bi was calculated from Eqs. (56) and (59). The extreme case, when $Bi \rightarrow \infty$ (the boundary condition of the first kind, Eq. (14)) was also considered. Such a case is very often applied in the literature. The moisture diffusion coefficient was calculated from Eq. (39) and by fitting Eq. (31) to the experimental data considering the lowest value of RMSE (Eq. (65)). As it was shown, the models of the first drying period (Eqs. (3), (9), and (11)) describe the course of drying curve for different range of time. Therefore Eq. (31) begins to model the second drying period in different moments and the values of the Biot number depend on the model applied for description of the first drying period. The various number of terms in analytical solution of Eq. (31) were taken into account. Moisture diffusion coefficients and the results of the statistical analyses are given in Table 2.

As can be seen from the statistical analysis results, the following model can be considered as the most appropriate: the model of the first drying period taking into account shrinkage (Eq. (11)) followed by the model of the second drying period for which moisture diffusion coefficient was calculated by fitting Eq. (31) to the experimental data considering the lowest value of RMSE. The mentioned model of the second drying period can be also considered as the most appropriate when the course of the drying curve at the second drying period is only taken under consideration. As the least appropriate for describing the course of the whole drying curve, the linear model of the first drying period followed by the model of the second drying period can be considered. It can be also noticed that the model of the second drying period for which moisture diffusion coefficient was calculated from Eq. (39) gives worse results comparing to model for which coefficient was calculated considering the lowest value of RMSE. Figure 3 presents the result of consistency verification of calculation results with empirical data. Analysis of obtained graph shows that results of calculations obtained from the discussed models are very well correlated with empirical data. The model of the first drying period taking into account shrinkage (Eq. (11)) is better correlated with empirical data comparing to model of the second drying period. Results of the statistical analyses (Table 1 and 2) confirm this regularity.

Model of the first drying period	Biot number Bi	Method of calculation of Bi	Number of terms in infinite series	Method of calculation of D	Moisture diffusion coefficient D	R^2 (for the second drying period)	RMSE (for the second drying period)	R^2 (for the whole drying process)	RMSE (for the whole drying process)
Eq. (3)	∞	-	10	Min(RMSE)	4.65×10^{-09}	0.986	0.2330	0.986	0.1901
			1		4.70×10^{-09}	0.994	0.2758	0.981	0.2247
	5.4	Eq. (56)	10	Eq. (39)	6.37×10^{-09}	0.991	0.1948	0.994	0.1592
			1			0.993	0.2107	0.992	0.1721
			10	Min(RMSE)	7.36×10^{-09}	0.991	0.1589	0.994	0.1303
			1		7.36×10^{-09}	0.996	0.1783	0.992	0.1460
	2.7	Eq. (59)	10	Eq. (39)	6.37×10^{-09}	0.982	0.3955	0.994	0.3218
			1			0.980	0.3971	0.992	0.3231
			10	Min(RMSE)	1.01×10^{-08}	0.994	0.1338	0.996	0.1101
			1		1.00×10^{-08}	0.996	0.1418	0.995	0.1166
Eq. (9)	∞	-	10	Min(RMSE)	3.01×10^{-11}	0.941	0.0464	0.999	0.0451
			1		3.19×10^{-11}	0.940	0.0479	0.999	0.0440
	0.07	Eq. (56)	10	Eq. (39)	9.51×10^{-10}	0.765	0.1970	0.999	0.0886
			1			0.765	0.1970	0.998	0.1064
			10	Min(RMSE)	8.92×10^{-09}	0.971	0.0332	0.999	0.0338
			1		9.10×10^{-09}	0.973	0.0331	0.999	0.0277
	0.04	Eq. (59)	10	Eq. (39)	9.51×10^{-10}	0.797	0.1624	0.999	0.0886
			1			0.797	0.1624	0.999	0.0886
			10	Min(RMSE)	5.44×10^{-09}	0.975	0.0344	0.999	0.0282
			1		5.48×10^{-09}	0.975	0.0344	0.999	0.0282
Eq. (11)	∞	-	10	Min(RMSE)	3.35×10^{-10}	0.992	0.0262	0.999	0.0207
			1		3.38×10^{-10}	0.973	0.0602	0.999	0.0269
	0.16	Eq. (56)	10	Eq. (39)	1.79×10^{-09}	0.867	0.2005	0.998	0.1149
			1			0.867	0.2005	0.998	0.1149
			10	Min(RMSE)	7.32×10^{-09}	0.992	0.0250	0.999	0.0233
			1		7.07×10^{-09}	0.991	0.0247	0.999	0.0233
	0.12	Eq. (59)	10	Eq. (39)	1.79×10^{-09}	0.848	0.2411	0.997	0.1377
			1			0.847	0.2411	0.997	0.1377
			10	Min(RMSE)	9.27×10^{-09}	0.991	0.0262	0.999	0.0238
			1		9.59×10^{-09}	0.992	0.0258	0.999	0.0238

Table 2. Moisture diffusion coefficients and the results of the statistical analyses

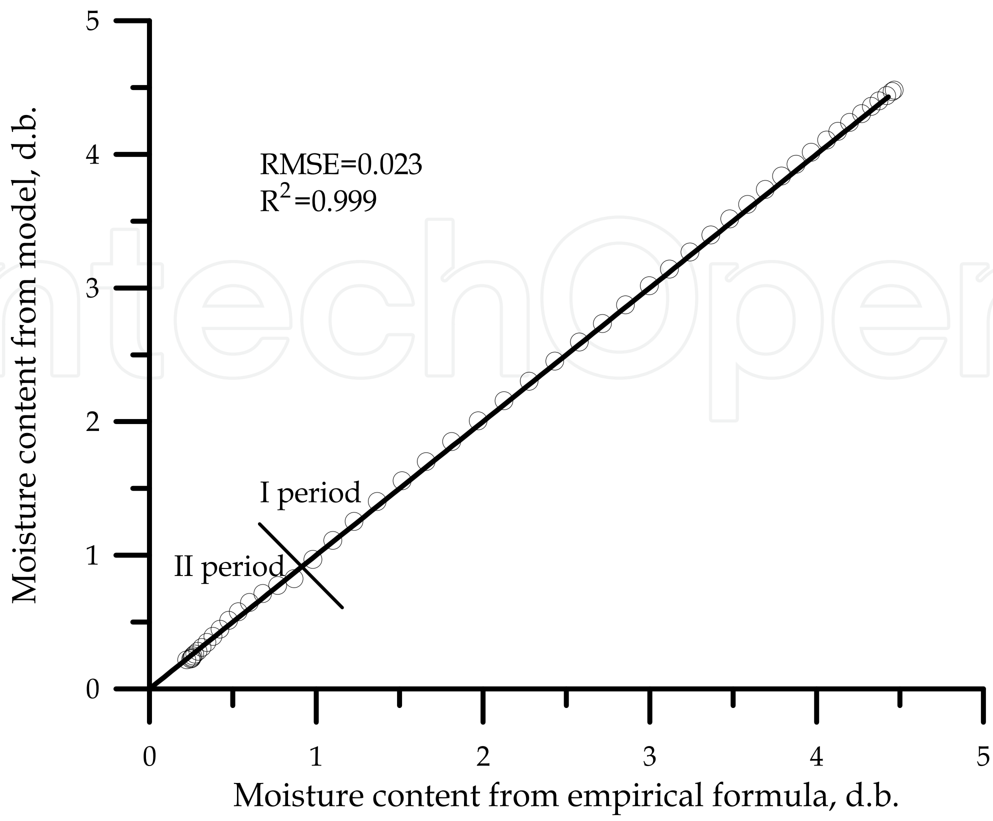

Fig. 3. Moisture content from model vs. experimental moisture content: I – first drying period, Eq. (11), II – second drying period, Bi=0.16, D from min(RMSE), 10 terms in infinite series

The determined moisture diffusion coefficient was found to be between $3.01{\cdot}10^{-11}$ m^2s^{-1} and $1.01{\cdot}10^{-8}$ m^2s^{-1} for the parsley root slices (Table 2). These values are within the general range for biological materials. Figures 4 and 5 show the influence of number of terms in infinite series in Eq. (31) on the value of obtained moisture diffusion coefficient and on the accuracy of verification of models of the second drying period. It can be accepted (Fig. 4) that the number of terms in infinite series do not influence much the value of the moisture diffusion coefficient. Its value was found to be between $3.33{\cdot}10^{-10}$ m^2s^{-1} and $3.41{\cdot}10^{-10}$ m^2s^{-1}. The influence of number of terms on RMSE was greater especially for number between i=1 (RMSE=0.06) and i=4 (RMSE=0.029). For higher number of terms the RMSE diminished very slowly and for i=10 reached the value of 0.026. Figure 5 presents the influence of number of terms in infinite series in Eq. (31) on the root mean square error RMSE and coefficient of determination R^2. The moisture diffusion coefficient determined for the first term in infinite series was then accepted in terms of higher number. It can be seen that the first four terms influence the accuracy of verification of Eq. (31) in higher degree than the next terms. The number of terms in Eq. (31) influences the obtained value of moisture ratio especially for values $0<Fo<0.08$, so in the beginning of the second drying period (Fig. 6). The first four terms influence the calculated moisture ratio in higher degree than the next terms. For values $Fo>0.08$, the solutions for various number of terms in infinite series are lying close together and truncating the series results in negligible errors.

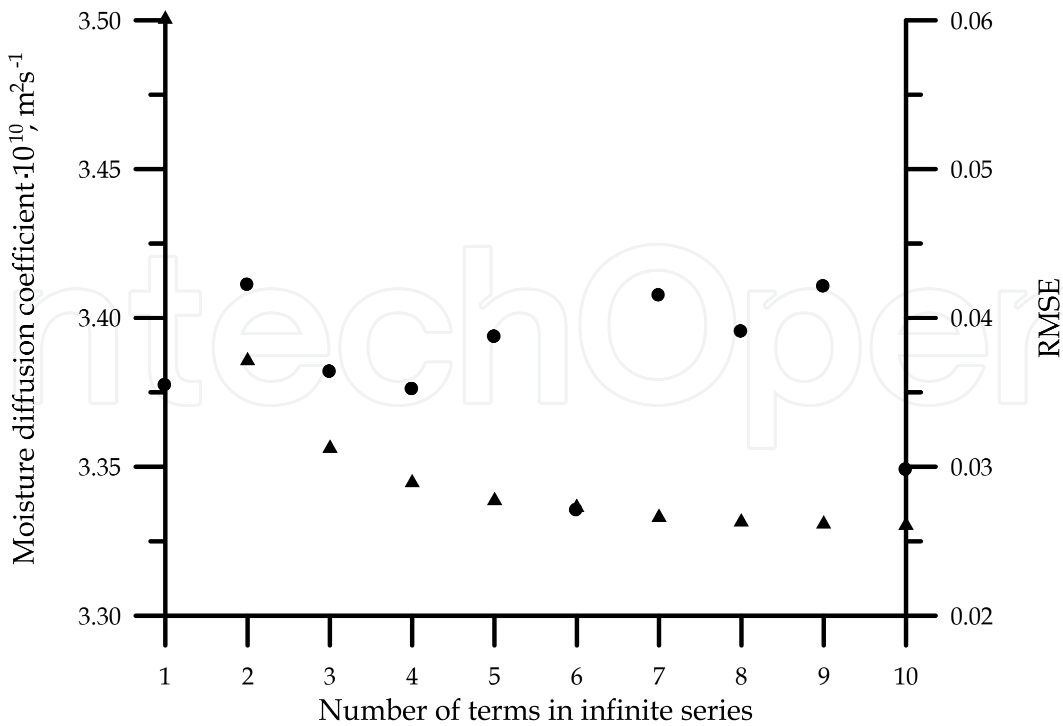

Fig. 4. Moisture diffusion coefficient vs. number of terms in infinite series in Eq. (31) and RMSE vs. number of terms in infinite series in Eq. (31) (first drying period – Eq. (11), Bi→∞): (●) – moisture diffusion coefficient, (▲) – RMSE

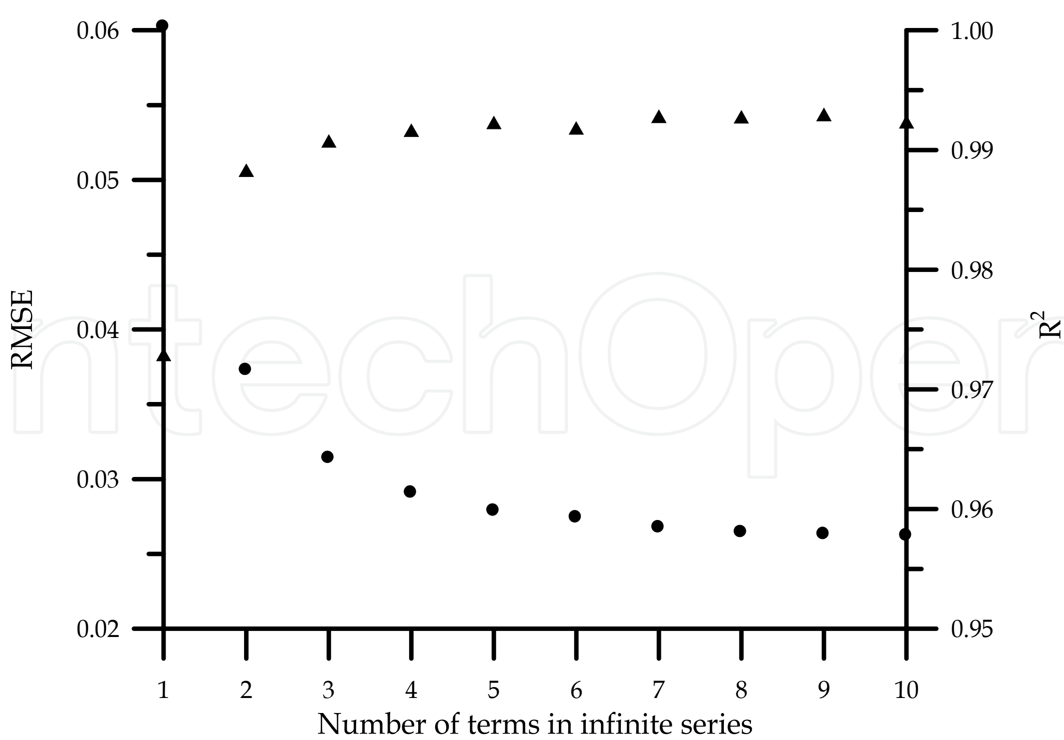

Fig. 5. RMSE vs. number of terms in infinite series in Eq. (31) and R^2 vs. number of terms in infinite series in Eq. (31) (first drying period – Eq. (11), Bi→∞): (●) – RMSE, (▲) – R^2

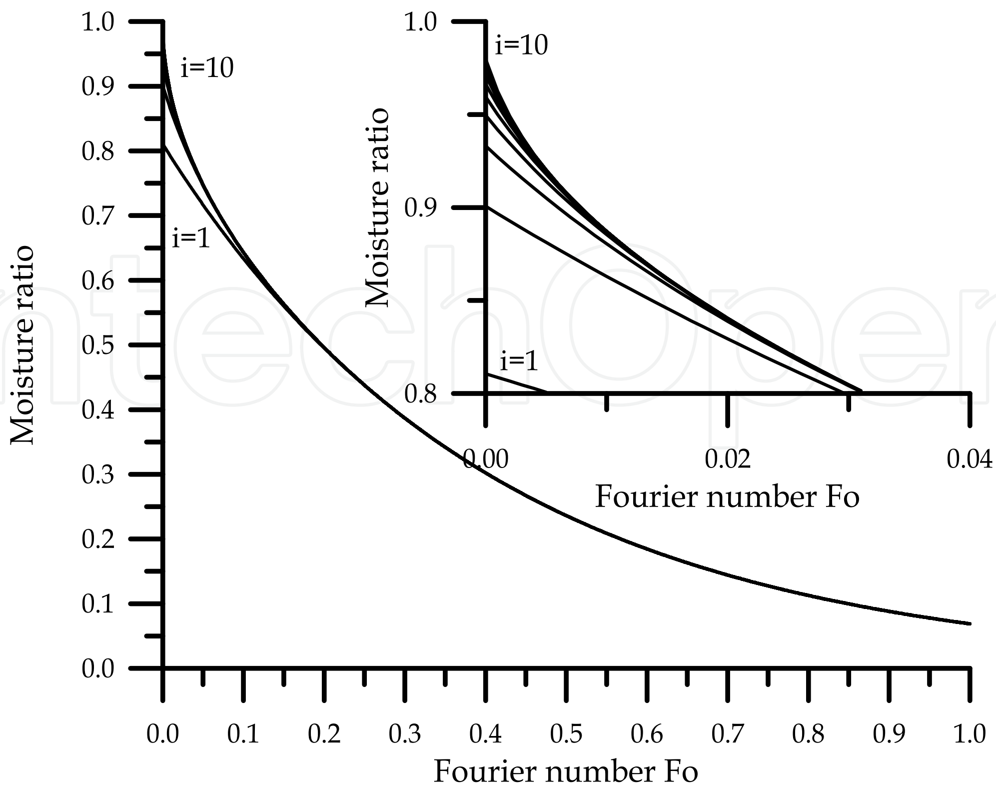

Fig. 6. Moisture ratio vs. Fourier number for various number of terms in infinite series in Eq. (31) (first drying period – Eq. (11), $Bi \to \infty$)

4. Conclusions

The results obtained from experiments and from mathematical model suggest that during the convective drying of parsley root slices there is a period of time during which the conditions of external mass transfer determine course of the process. The results of the linear model Eq. (3) verification indicate that during the drying of parsley root slices the period of constant drying rate takes place. Verified models of the first drying period Eqs. (9) and (11) taking into account drying shrinkage confirm that the decrease of the drying rate during the first drying period of parsley root slices can be caused by the shrinkage of drying slices.
Model of infinite plane drying accurately predicts the drying curve in the second drying period for parsley root slices. The determined moisture diffusion coefficient was found to be between $3.01 \cdot 10^{-11}$ m^2s^{-1} and $1.01 \cdot 10^{-8}$ m^2s^{-1}. These values are within the general range for biological materials. The number of terms in model of infinite plane drying influences the obtained solution especially in the beginning of the second drying period.
The course of the whole drying curve for parsley root slices could be described satisfactorily by using only the model with drying shrinkage. This model do not explain, however, the phenomenon of drying in the second period therefore applying such a model to the whole drying curve has only practical meaning.

5. Nomenclature

A surface area of dried solid (m^2)
a,b constants (Eqs. (35), (55), and (63))

a, b, c constants (Eqs. (36), (37), (49), (50), (52), (53), (54), and (62))
a, b, c, d constants (Eq. (51))
A_0 initial surface area of dried solid (m^2)
Ar Archimedes number ($Ar=gR^3\Delta\rho/\nu^2\rho$)
A_w the part of surface A on which mass flux is not equal to zero (m^2)
b dimensionless empirical coefficient of shrinkage model (Eq. (5))
Bi Biot number ($Bi=h_mR/D$)
c specific heat (J kg^{-1} K^{-1})
D moisture diffusion coefficient (effective diffusivity) (m^2 s^{-1})
D_0 diffusion coefficient at the reference temperature (m^2 s^{-1})
D_{wa} diffusion coefficient of water vapour (m^2 s^{-1})
E_a activation energy (J mol^{-1})
Fo Fourier number ($Fo=Dt/R^2$)
g acceleration of gravity (m s^{-2})
G lag factor
Gr Grashof number ($Gr=gR^3\beta\Delta T/\nu^2$)
Gr_m Grashof number (mass) ($Gr_m=gR^3\beta'\Delta p/\nu^2$)
h half of cylinder height (m)
h heat transfer coefficient ($Wm^{-2}K^{-1}$)
h_m mass transfer coefficient (m s^{-1})
k initial drying rate (s^{-1})
k_{th} thermal conductivity (Wm^{-1} K^{-1})
L latent heat of water vaporization (J kg^{-1})
M moisture content (dry basis)
M_0 initial moisture content (dry basis)
M_c critical moisture content (dry basis)
M_e equilibrium moisture content (dry basis)
MR dimensionless moisture content, moisture ratio
MR_{exp} experimental moisture ratio
MR_{pre} predicted moisture ratio
N dimensionless empirical coefficient (Eq. (11))
Nu Nusselt number ($Nu=hR/k_{th}$)
n dimensionless empirical coefficient of shrinkage model (Eq. (8))
n orthogonal to surface A
n_1 dimensionless empirical coefficient (Eq. (10))
p pressure (Pa)
Pr Prandtl number ($Pr=\nu/\alpha$)
R universal gas constant (J mol^{-1} K^{-1})
R characteristic dimension (m)
R_1, R_2, R_3 half of cube thickness (m)
R_c half of plane thickness or cylinder radius (m)
Re Reynolds number ($Re=uR/\nu$)
RMSE root mean square error
R^2 coefficient of determination
r, x, y, z coordinates (m)
S drying coefficient (s^{-1})
Sc Schmidt number ($Sc=\nu/D_{wa}$)

Sh Sherwood number ($Sh=h_mR/D_{wa}$)
T temperature (°C)
T_a temperature of drying air (°C)
T_A temperature of solid surface (°C)
T_{wb} wet-bulb temperature (°C)
t time (s)
t_c time of drying while moisture content $M=M_c$ (s)
u velocity (m s^{-1})
V volume of the dried solid (m^3)
V_0 initial volume of the dried solid (m^3)
V_s volume of the dry matter (m^3)
W_s dry matter of solid (kg)
$\dot{W}$ mass flux (kg $m^{-2}s^{-1}$)
α thermal diffusivity (m^2 s^{-1})
β volumetric expansion coefficient (K^{-1})
β' coefficient (m^2 N^{-1})
ν kinematic viscosity (m^2 s^{-1})
ρ_s density of dry matter (kg m^{-3})
Δ increment

5.1 Subscripts
A outer surface of body

5.2 Superscripts
$\bar{}$ average value in volume V

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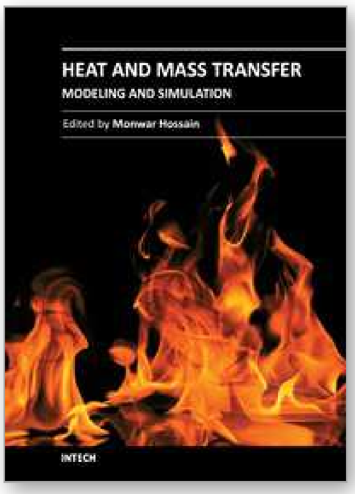

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