

## MOISTURE DIFFUSIVITY ESTIMATION FROM TEMPERATURE MEASUREMENTS: INFLUENCE OF MEASUREMENT ACCURACY

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### ABSTRACT

In this paper a method of moisture diffusivity estimation on the basis of a drying body temperature response is analyzed by using an inverse approach. The method takes advantage of the interrelation between the heat and mass transport processes within the drying body as well as from its surface to the surroundings. As a result, the low accuracy local moisture content or standard drying curve measurements can be substituted by this accurate and easy to perform method that requires only single thermocouple temperature measurements. The Levenberg-Marquardt procedure is applied for estimation of the moisture content and temperature dependent moisture diffusivity. Numerical experiments have been conducted to investigate sensitivity of this method to the heat and mass transfer coefficients accuracy. In order to simulate real measurements, a normally distributed error was added to the numerical temperature response. An analysis of the influence of the drying air velocity, drying air temperature, drying body dimension and drying time on the moisture diffusivity estimation that enables the design of the proper experiment is presented as well.

### INTRODUCTION

Inverse approach to parameter estimation in last few decades has become widely used in various scientific domains. While in the classical direct problem the cause is given and the effect is determined, the inverse problem involves estimation of the cause from the knowledge of the effect. The solution of

inverse problems often requires the solution of their direct problems. Therefore, the ability of an inverse problem method is often closely related to direct problem method of solution.

There are several methods for describing direct problem of the complex simultaneous heat and moisture transport processes within drying material. In the approach proposed by Luikov (1972) the moisture and temperature fields in the drying body are expressed by a system of two coupled partial differential equations. The system of equations incorporates coefficients, which are functions of temperature and moisture content, and must be determined experimentally. For many practical calculations the influence of the temperature and moisture content on all the transport coefficients except for the moisture diffusivity is small and can be neglected.

For many drying processes, including the drying processes considered in this paper, the influence of the thermal diffusion is small and can be ignored. In this case, the Luikov's moisture transport equation is the same as the Fick's second law equation, where concentration has been converted to moisture content on a dry basis. The moisture diffusivity has the same meaning in both of these approaches. It accounts for various types of possible drying processes including molecular (liquid) diffusion, vapor diffusion, surface diffusion, hydrodynamic flow, Knudsen flow, and other considerations. An effective moisture diffusivity, which lumps all possible moisture transport mechanisms into a single measurable parameter, is often used to characterize the drying behavior regardless of the dominating mechanism (Feng et al., 1999). The moisture diffusivity dependence on moisture content and temperature exerts a strong influence on the drying process calculation. This effect can not be ignored for the majority of practical cases.

All the coefficients except for the moisture diffusivity can be relatively easily determined by experiments (Karathanos et al., 1996; Rahman, 1995). A number of methods for the experimental determination of the moisture diffusivity exist such as: sorption kinetics methods, permeation methods, concentration-distance methods, drying methods, radiotracer methods, and methods based on the techniques of electron spin resonance and nuclear magnetic resonance. There is no standard method for the experimental determination of the moisture diffusivity. The adoption of a generalized method for moisture diffusivity estimation would be of great importance; however, this does not seem probable in the near future (Zogzas et al., 1996).

The application of the moisture diffusivity estimation methods based on the experimental drying curves in relation to the analytical solution of the differential diffusion equation seems to be the most popular experimental practice (Zogzas and Maroulis, 1996; Feng et al., 1999). Numerical solutions of the Fick's law differential diffusion equation with constant (Daud et al., 1997) or moisture and temperature dependent (Zogzas and Maroulis, 1996) diffusivity also have been used for the moisture diffusivity estimation.

The main idea of the present method is to make use of the interrelation between the heat and mass (moisture) transport processes within the drying body and from its surface to the surroundings. Then, the moisture diffusivity can be estimated on the basis of an accurate and easy to perform single thermocouple temperature measurement by using an inverse approach. Kanevce, Kanevce and Dulikravich (2000a, 2000b) and Dantas et al. (2000) recently analyzed this idea of the moisture diffusivity estimation by temperature response of a drying body.

In this paper, the solution of the inverse problem of estimating the moisture content and temperature-dependent moisture diffusivity is presented. The present parameter estimation problem is solved by using the Levenberg-Marquardt method of minimization of the least-squares norm, by using simulated experimental data with random errors. Instead of actual temperature measurements, the temperature response during convective drying is obtained from the numerical solution of the non-linear one-dimensional Luikov's equations. In order to simulate real measurements, a normally distributed error was added to the numerical temperature response. As a representative drying body, a mixture of bentonite and quartz sand with known thermophysical properties has been chosen.

The objective of this paper is an analysis of the sensitivity of this method of moisture diffusivity estimation to the heat and mass transfer coefficients accuracy. An analysis of the influence of the drying air velocity, drying air temperature, drying body dimension and drying time on the moisture diffusivity

estimation that enables the design of the proper experiment is presented as well. In order to realize this analysis the sensitivity coefficients and the sensitivity matrix determinant were calculated (Kanevce, Kanevce and Dulikravich, 2000b) for the characteristic drying regimes and drying body dimensions.

## MATHEMATICAL MODEL OF DRYING

In the case of an infinite flat plate of thickness  $2L$ , if the shrinkage of the material during drying can be neglected, the resulting system of equations for the temperature,  $T(x, t)$ , and moisture content,  $X(x, t)$ , can be expressed as

$$c\rho_s \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( k \frac{\partial T}{\partial x} \right) + \varepsilon \rho_s \Delta H \frac{\partial X}{\partial t} \quad (1)$$

$$\frac{\partial X}{\partial t} = \frac{\partial}{\partial x} \left( D \frac{\partial X}{\partial x} + D\delta \frac{\partial T}{\partial x} \right) \quad (2)$$

Here,  $t$ ,  $x$ ,  $c$ ,  $k$ ,  $\Delta H$ ,  $\varepsilon$ ,  $\delta$ ,  $D$ ,  $\rho_s$  are time, distance from the mid-plane of the plate, heat capacity, thermal conductivity, latent heat of vaporization, ratio of water evaporation rate to the reduction rate of the moisture content, thermo-gradient coefficient, moisture diffusivity, and density of the dry plate material, respectively.

As initial conditions, uniform temperature and moisture content profiles are assumed

$$t = 0 \quad T(x,0) = T_0, \quad X(x,0) = X_0 \quad (3)$$

The boundary conditions on the free plate surface ( $x = L$ ) are

$$\begin{aligned} -k \left( \frac{\partial T}{\partial x} \right)_{x=L} + j_q - \Delta H(1 - \varepsilon)j_m &= 0 \\ D\rho_s \left( \frac{\partial X}{\partial x} \right)_{x=L} + D\delta\rho_s \left( \frac{\partial T}{\partial x} \right)_{x=L} + j_m &= 0 \end{aligned} \quad (4)$$

In the case of convective drying of the sample, the convective heat flux,  $j_q(t)$ , and mass flux,  $j_m(t)$ , on the surface of evaporation are

$$\begin{aligned} j_q &= h(T_a - T_{x=L}) \\ j_m &= h_D(C_{x=L} - C_a) \end{aligned} \quad (5)$$

where  $h$  is the heat and  $h_D$  the mass transfer coefficient,  $T_a$  is the drying air bulk temperature, and  $C_a$  is the concentration of water vapor in the drying air. The water vapor concentration of the air in equilibrium with the free surface of the body is calculated by

$$C_{x=L} = a(T_{x=L}, X_{x=L}) \cdot p_s(T_{x=L}) / 461.9 / (T_{x=L} + 273) \quad (6)$$

Here  $p_s$  is the saturation pressure and  $a$  is the water activity calculated from the water sorption isotherms. The problem is symmetrical, and boundary conditions on the symmetry surface of the plate are

$$\left(\frac{\partial T}{\partial x}\right)_{x=0} = 0, \quad \left(\frac{\partial X}{\partial x}\right)_{x=0} = 0 \quad (7)$$

The system of equations (1) and (2) with the initial (3) and the boundary conditions (4) and (7) has been solved numerically. In order to approximate the solution an explicit procedure has been used (Kanevce, Kanevce and Dulikravich, 2000a).

## MOISTURE DIFFUSIVITY ESTIMATION

The proposed method of the moisture diffusivity estimation by temperature response of a drying body was tested for a model material (Kanevce, Kanevce and Dulikravich, 2000a) which was a mixture of bentonite and quartz sand with known thermophysical properties (Kanevce et al., 1980; Kanevce, 1998). From the experimental and numerical examinations of the transient moisture and temperature profiles (Kanevce, 1998) it was concluded that for the calculations in this study, the influence of the thermal diffusion is small and can be ignored. It was also concluded that the Luikov's system of simultaneous partial differential equations could be used by treating the transport coefficients as constants except for the moisture diffusivity. The appropriate mean values for the model material are: the density of dry solid,  $\rho_s = 1738 \text{ kg/m}^3$ , heat capacity,  $c = 1550 \text{ J/(kgK)}$ , thermal conductivity,  $k = 2.06 \text{ W/(mK)}$ , latent heat of water vaporization,  $\Delta H = 2.31 \cdot 10^6 \text{ J/kg}$ , ratio of water evaporation rate to the reduction rate of the moisture content,  $\varepsilon = 0.5$  and thermo-gradient coefficient,  $\delta = 0$ . The experimentally obtained desorption isotherms of the model material are presented by the empirical equation

$$a = 1 - \exp(-1.5 \cdot 10^6 \cdot (T + 273)^{-0.91} \cdot X^{-0.005 \cdot (T+273)+3.91}) \quad (8)$$

Any other type of equation, corresponding to the Smith, BET, GAB ... model can be used in the presented method.

The following empirical function of temperature and moisture content can describe the experimentally obtained relationship for the moisture diffusivity (Kanevce et al., 1980)

$$D = \frac{D_1}{D_2 + X^2} \cdot \left(\frac{T + 273}{303}\right)^{10} \quad (9)$$

where  $D_1$  and  $D_2$  are constants. The values for the model material are:  $D_1 = 9.0 \times 10^{-12}$  and  $D_2 = 0$ . Arrhenius-type equation for the moisture diffusivity can be also used in the presented method.

For the inverse problem investigated here, values of  $D_1$  and  $D_2$  are regarded as unknown parameters and all other quantities figuring in direct problem formulation were assumed to be known. For the estimation of these parameters, we consider available the transient single thermocouple temperature measurements. The estimation methodology used is based on minimization of the ordinary least square norm

$$E(P) = [Y - T(P)]^T [Y - T(P)] \quad (10)$$

Here,  $Y^T = [Y_1, Y_2, \dots, Y_M]$  is a vector of measured temperatures and  $T = [T_1(P), T_2(P), \dots, T_M(P)]$  is a vector of estimated temperatures at the measurement location at time  $t_i$  ( $i = 1, 2, \dots, M$ ), while  $P^T = [P_1, P_2, \dots, P_N]$  is the vector of unknown parameters,  $M$  is total number of measurements, and  $N$  is the total number of unknown parameters ( $M \geq N$ ).

A version of Levenberg-Marquardt method was applied for the solution of the presented parameter estimation problem (Marquardt, 1963). This method is quite stable, powerful, and straightforward. It has been applied to a variety of inverse problems and belongs to damped least square methods (Beck and Arnold, 1977). The solution for P is achieved using the following iterative procedure

$$P^{r+1} = P^r + [(J^r)^T J^r + \mu^r I]^{-1} (J^r)^T [Y - T(P^r)] \quad (11)$$

where I is the identity matrix,  $\mu$  is the damping parameter, and J represents sensitivity matrix.

$$J = \begin{bmatrix} \frac{\partial T_1}{\partial P_1} & \dots & \frac{\partial T_1}{\partial P_N} \\ \vdots & & \vdots \\ \frac{\partial T_M}{\partial P_1} & \dots & \frac{\partial T_M}{\partial P_N} \end{bmatrix} \quad (12)$$

The term  $\mu I$  damps instabilities due to ill-conditioned character of the problem. Near the initial guess, the problem is generally ill-conditioned and damping parameter is chosen large making term  $\mu I$  large as compared to term  $J^T J$ . So, the matrix  $J^T J$  is not required to be non-singular at the beginning of iterations and the procedure tends towards a slow-convergent steepest descent method. As the iteration process approaches the converged solution, the damping parameter decreases, and the Levenberg-Marquardt method tends towards Gauss method. In fact, this method compromises between the steepest descent and Gauss method choosing  $\mu$  so as to follow the Gauss method to as large an extent as possible, while retaining a bias towards the steepest descent direction to prevent instabilities. The presented iterative procedure stops if the norm of gradient of E(P) is sufficiently small, or if the ratio of the norm of gradient of E(P) to the E(P) is small enough, or if the changes in the vector of parameters are very small.

## RESULTS AND DISCUSSION

The presented method allows a moisture and temperature dependent heat and mass transfer coefficients to be used. The best approach should be if the appropriate relations are obtained by comparison of the known with individual experiments realized on the same experimental set-up for the moisture diffusivity estimation. In this case the large value for the mass transfer Biot number indicates that the internal resistance controls the drying process. Consequently, the moisture content of the surface of the body is practically constant, near equilibrium moisture content, very short after the beginning of the drying (Figure 4). In conclusion, constant heat and mass transfer coefficients are taken for the purposes of this analysis. The corresponding mean values have been calculated from the Nesterenko's relations (Luikov, 1972) for heat and mass Nusselt numbers in drying conditions.

In order to investigate influence of the boundary conditions, determinant of the sensitivity matrix  $J^T J$  with normalized elements

$$[J^T J]_{m,n} = \sum_{i=1}^M \left( P_m \frac{\partial T_i}{\partial P_m} \right) \left( P_n \frac{\partial T_i}{\partial P_n} \right), \quad m, n = 1, \dots, N \quad (13)$$

and the relative sensitivity coefficient,  $D_1 \partial T_i / \partial D_1$ ,  $i = 1, 2, \dots, M$ , were calculated (Kanevce, Kanevce, Dulikravich, 2000b). The sensitivity coefficients analysis was carried out for the plate of thickness  $2L$ , with initial moisture content of  $X(x, 0) = 0.20$  kg/kg and initial temperature  $T(x, 0) = 20$  °C. In order to

investigate the influence of the boundary conditions, the drying air bulk temperature,  $T_a$ , and velocity,  $V_a$ , were perturbed. The relative humidity of the drying air was  $\varphi = 0.12$ . The test cases are shown in the Table 1.

Table 1. Drying air conditions

Thickness 2L		Drying air		Transfer coefficients	
3 mm	6 mm	$T_a$ [ $^{\circ}$ C]	$V_a$ [m/s]	$h$ [W/m $^2$ K]	$h_D 10^2$ [m/s]
A1	AA1	80	3	28.6	3.20
B1	BB1	80	5	44.9	5.02
C1	CC1	80	10	83.1	9.29
A2	AA2	120	3	29.3	3.55
B2	BB2	120	5	45.9	5.56
C2	CC2	120	10	84.9	10.30

Table 1 also contains the corresponding convective heat, and mass transfer coefficients  $h$  and  $h_D$ , obtained for an 80 mm long plate. The test cases were repeated for two different thicknesses of the drying body,  $2L = 3$  mm and 6 mm.

Figure 1 shows the determinant of the sensitivity matrix  $J^T J$  with normalized elements and Figure 2 the relative sensitivity coefficient for all test cases depicted in Table 1. Since the sample drying object represented by a flat plate is very thin, a single thermocouple was located in the mid-plane of the infinite flat plate. Two plateaus can be seen on the presented sensitivity determinant curves. The first plateau corresponds to the moment (Kanevce, Kanevce, Dulikravich, 2000a) when the body moisture content is nearly equal to the equilibrium. After that, small evaporation rate and fast drying rate decrease accompanied by fast body temperature increase occur. The second plateau corresponds to the moment when nearly equilibrium temperature was obtained.

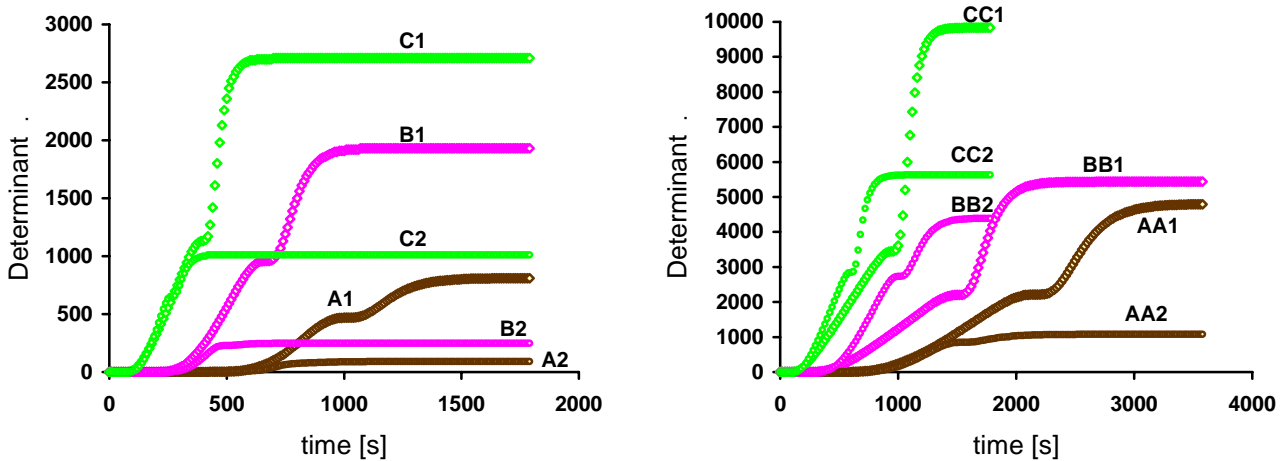


Figure 1. Sensitivity Determinants

The tendency of increasing determinant as well as sensitivity coefficient with drying air velocity can be explained by the increasing of the mass transfer Biot number and the moisture content gradients inside the body. The heat transfer Biot number and the temperature gradients (Figure 4) are very small in all the cases. The mass transfer Biot number changes during the drying process as a function of the local moisture content and temperature change. The value of this Biot number is very high, practically infinite, at the beginning of the drying, and tends to low values at the end of the drying. Under these conditions

mostly the moisture diffusivity and dimensions of the body govern the process of drying. The same conclusion remains when plate thickness increase. The influence of the drying air temperature also can be explained through the mass transfer Biot number and the moisture content gradients inside the body. Higher drying air temperature leads to a higher drying body temperature and lower moisture content (higher drying rate) that lead to higher moisture diffusivity, and consequently lower mass transfer Biot number. That leads to a tendency of decreasing determinant and sensitivity coefficient with increase in drying air temperature.

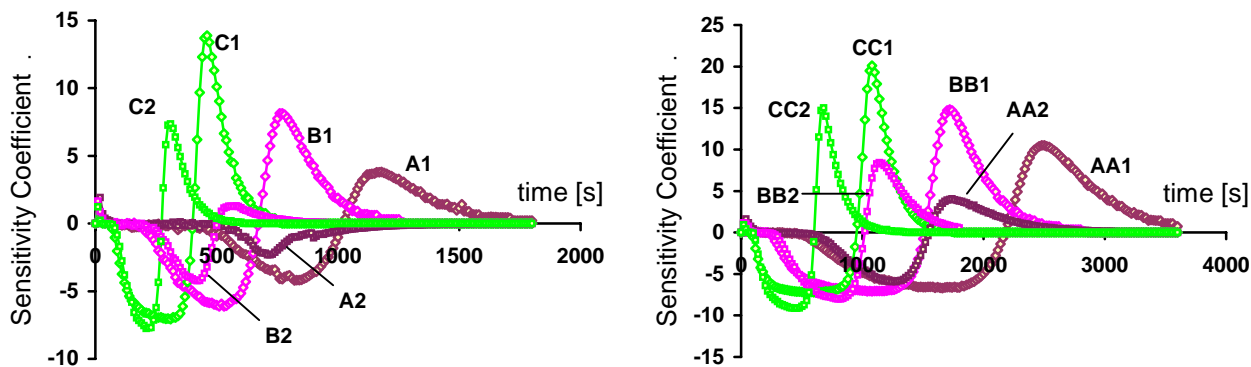


Figure 2. Sensitivity Coefficients

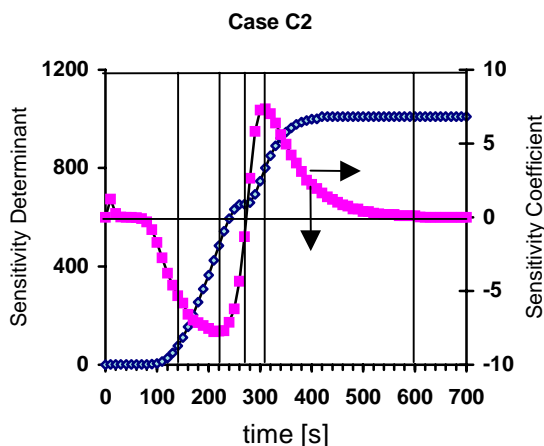


Figure 3. The Characteristic Times

2000b) for the test case (C2). Five drying times, corresponding to (Figure 3 and Figure 4) were considered: the end of drying and the maximum determinant value (600 s), the maximum sensitivity coefficient value (310 s), the first determinant value plateau and zero sensitivity coefficient value (272 s), the maximum negative sensitivity coefficient value (220 s), and the drying time below that (140 s). Very good agreement was achieved for all the cases with drying time equal or longer than that corresponding to the first plateau on the determinant curve. The results showed the tendency of increasing  $D_1$  accuracy with determinant. The mean accuracy of  $D_2$  was very high for all the cases. For the drying times shorter than that corresponding to the first plateau, the problem is ill-posed and local minimums were obtained depending on the initial guesses.

For the estimation of the moisture diffusivity available the transient readings of the temperature sensor located in the mid-plane have been considered. Instead of actual temperature measurements, the temperature response during convective drying is obtained from the numerical solution of the non-linear one-dimensional Luikov's equations, by treating the values and expressions for the material properties as known. In order to simulate real measurements, a normally distributed error with zero mean and standard deviation of 0.5 °C was added to the numerical temperature response.

In order to investigate influence of the considered duration of temperature response parameters  $D_1$  and  $D_2$  were estimated (Kanevce, Kanevce, Dulikravich,

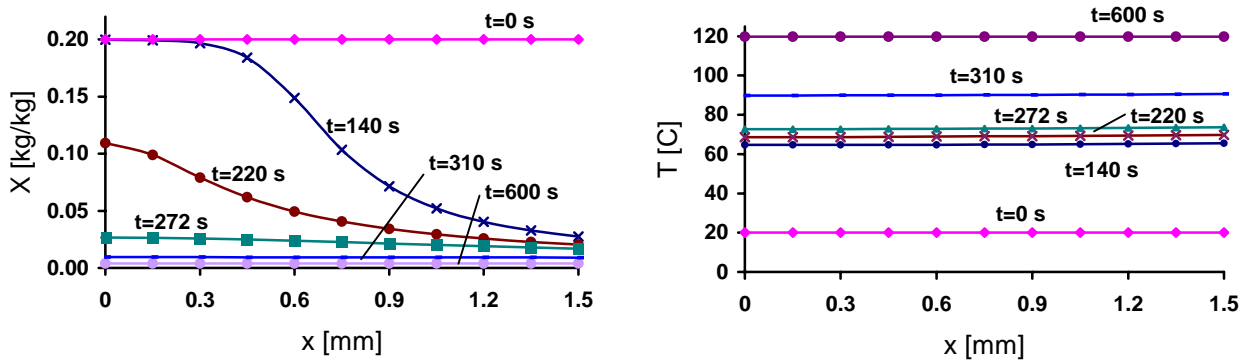


Figure 4. Transient Moisture Content and Temperature Profiles (Case C2)

In order to investigate influence of heat and mass transfer coefficients perturbations on the moisture diffusivity estimation accuracy, test cases presented in Table 2 have been analyzed. Case C2 has been chosen owing to the shortest drying time and case CC1 because of its highest values of sensitivity coefficient and determinant. The selected characteristic drying times enable accurate estimating of the moisture diffusivity if "exact" values for heat and mass transfer coefficients are used as input data. The temperature response had constant time-step of 2 s, so different number of measured temperatures has been taken for the cases with different drying time. The number of the space grid points has been 41 in all the drying process calculation schemes. The initial guess for all the cases was the same and it was very far from the exact values of parameters :  $D_{1init} = 5.0 \times 10^{-11}$  and  $D_{2init} = 0.1$ .

To calculate moisture diffusivity values in Table 2, besides the "exact" values, 10% increased values for one or both heat and mass transfer coefficients were used simulating data with measurement errors. For the comparison, the exact parameter values are also shown. The relative error has been calculated as  $\epsilon_1(\%) = 100 \cdot |(D_1 - D_{1ex}) / D_{1ex}|$ . The  $D_2$  accuracy depends on the  $X^2$  value and is changing from the beginning to the end of drying,  $\epsilon_2 = ((X^2 + D_2) - X^2) / X^2 = D_2 / X^2$ . The mean relative error of  $D_2$  has been calculated with the mean moisture content value  $X_m$  as  $\epsilon_2(\%) = 100 \cdot |D_2 / X_m^2|$ . As a mean moisture content value during the drying,  $X_m = 0.10$  kg/kg has been used for all the cases.

Table 2. Influence of heat and mass transfer coefficients' perturbations

Case	t[s]	h[W/m <sup>2</sup> K]	h <sub>D</sub> 10 <sup>2</sup> [m/s]	D <sub>1</sub> ·10 <sup>12</sup>	ε <sub>1</sub> [%]	D <sub>2</sub>	ε <sub>2</sub> [%]
C2	600	84.9	10.30	9.01	0.13	8.30·10 <sup>-7</sup>	8.0·10 <sup>-3</sup>
C2	272	84.9 x 1.10	10.30 x 1.10	6.96	29.3	1.04·10 <sup>-1</sup>	1.0·10 <sup>3</sup>
C2	272	84.9	10.30 x 1.10	8.50	5.9	1.17·10 <sup>-1</sup>	1.2·10 <sup>3</sup>
C2	600	84.9	10.30 x 1.10	8.72	3.2	4.79·10 <sup>-6</sup>	4.8·10 <sup>-2</sup>
C2	600	84.9 x 1.10	10.30	8.64	4.2	4.15·10 <sup>-4</sup>	4.1
C2	600	84.9 x 1.10	10.30 x 1.10	8.39	7.3	3.81·10 <sup>-4</sup>	3.8
CC1	1800	83.1 x 1.10	9.29 x 1.10	8.30	8.4	5.54·10 <sup>-5</sup>	0.55
Exact values				9.00		0.00	

It can be seen that for the drying time of 272 s for Case C2 the obtained results are significantly different from exact values, especially for  $D_2$ . The other test cases show that perturbations of heat and mass transfer coefficients produce reduced errors of estimated moisture diffusivity parameters if whole duration of drying is taken in the analysis.



## CONCLUSIONS

The method for estimation of moisture diffusivity on the basis of thermal transient response of a drying body by using inverse approach is presented. The Levenberg-Marquardt method is applied for evaluation of unknown parameters in moisture diffusivity dependence on moisture and temperature. The results obtained with simulated measurements show good agreement between evaluated and exact parameter values and confirm the validity of the proposed method.

Tendency of increasing determinant as well as sensitivity coefficient was obtained with drying air velocity and drying body thickness increase and with drying air temperature decrease. This can be explained by the increase of the mass transfer Biot number and the moisture content gradients inside the body. The heat transfer Biot number and the temperature gradients are very small. Under these conditions, mostly the moisture diffusivity and dimensions of the body govern the process of drying.

The influence of the drying air velocity increasing on the determinant and drying time leads to the computational time decreasing. The opposite influence of the drying air temperature and drying body thickness on the determinant and drying time suggests that additional research needs to be done in defining its optimal values concerning the minimal computational time.

Concerning the sensitivity of this method to the heat and mass transfer coefficient accuracy, it can be concluded that perturbations (simulated errors) in heat and mass transfer coefficients produce reduced errors of estimated moisture diffusivity parameters.

## NOTATION

a	water activity	-
c	heat capacity	J/K/kg db
C	concentration of water vapor in air	kg/m <sup>3</sup>
D	moisture diffusivity	m <sup>2</sup> /s
h	heat transfer coefficient	W/m <sup>2</sup> /K
h <sub>D</sub>	mass transfer coefficient	m/s
I	identity matrix	-
j <sub>m</sub>	mass flux	kg/m <sup>2</sup> s
j <sub>q</sub>	heat flux	W/m <sup>2</sup>
J	sensitivity matrix	-
k	thermal conductivity	W/m/K
L	flat plate thickness	m
p <sub>s</sub>	saturation pressure	Pa
P	vector of unknown parameters	-
t	time	s
T	temperature	<sup>0</sup> C
<b>T</b>	vector of estimated temperature	<sup>0</sup> C
V	velocity	m/s
x	distance from the mid-plane	m
X	moisture content (dry basis)	kg/kg db
Y	vector of measured temperature	<sup>0</sup> C
<i>Greek Symbols</i>		-
δ	thermo-gradient coefficient	1/K
ΔH	latent heat of vaporization	J/kg
ε	water evaporation rate ratio	-
μ	damping parameter	-
ρ	density	kg/m <sup>3</sup>
φ	relative humidity	-

*Subscripts*

a        air  
s        dry solid

*Superscripts*

r        iteration number  
T        transpose

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