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INFLUENCE OF BOUNDARY CONDITIONS ON MOISTURE DIFFUSIVITY ESTIMATION BY TEMPERATURE RESPONSE OF A DRYING BODY

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ABSTRACT
The objective of this paper is 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. The main idea of the presented method is to use the interrelation between the heat and moisture transport processes within the drying body for the moisture diffusivity estimation on the basis of a temperature response. The moisture and temperature fields in the drying body are described by a system of two coupled partial differential equations. All the coefficients except for the moisture diffusivity are taken as known. The Levenberg-Marquardt inverse approach is applied for the estimation of the moisture content and temperature-dependent moisture diffusivity. The temperature responses during convective drying of a representative body with known properties have been obtained by simulated experiments. The sensitivity coefficients and the sensitivity matrix determinant have been calculated for the characteristic regimes in order to define the influence of the drying air velocity and temperature, drying body dimension, and drying time. A correlation between the mass transfer Biot number and the sensitivity coefficient and determinant is established.

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NOMENCLATURE

a = water activity
c = heat capacity, *J/K/kg db*
C = concentration of water vapor in air, *kg/m*^{3}
D = moisture diffusivity, *m*^{2}*/s*
h = heat transfer coefficient, *W/m*^{2}*/K*
h_D = mass transfer coefficient, *m/s*
ΔH = latent heat of vaporization, *J/kg*
I = identity matrix
j_m = mass flux, *kg/m*^{2} *s*
j_q = heat flux, *W/m*^{2}
J = sensitivity matrix
κ = thermal conductivity, *W/m/K*
L = flat plate thickness, *m*
p_s = saturation pressure , *Pa*
P = vector of unknown parameters
t = time, *s*
T = temperature, ^0C
T = vector of estimated temperature, ^0C
V = velocity, *m/s*
x = distance from the mid-plane, *m*
X = moisture content (dry basis), *kg/kg db*
Y = vector of measured temperature, ^0C
δ = thermo-gradient coefficient, *1/K*
ε = ratio of water evaporation rate to the reduction rate of the moisture content
ε = relative error

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ε = relative error
σ = standard deviation
μ = damping parameter
ρ = density, kg/m^3
φ = relative humidity

Subscripts
a = drying air
s = dry solid

INTRODUCTION

There are several different methods of describing the complex simultaneous heat and moisture transport processes within drying material. In the approach initially proposed by Philip and De Vries (1957) and Luikov (1968) the moisture and temperature fields in the drying body are described by a system of two coupled partial differential equations. Strictly speaking, beside the temperature and water content, the gaseous pressure should be used as an independent variable. But the assumption of a uniform gaseous pressure has been widely accepted for a variety of specific applications (Mukherjee et al., 1997; Bastian, 1997). The system of equations incorporates coefficients that are functions of temperature and moisture content and is non-linear. For some applications the non-linear system of two coupled partial differential equations have been used (Sadykov, 1998; Bastian, 1997; Kanevce, 1998), but, for many practical calculations, the influence of the temperature and moisture content on all the transport coefficients has been neglected and the system of linear equations has been used (Chang, 1998; Dantas, 1999; Gong, 1998; Mukherjee et al., 1997; Songtao et al., 1999).

For many drying processes, the influence of the thermo-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 (Zogzas et al., 1994) 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; Ramaswamy and Nsonzi, 1998; Markowski, 1998; Ruiz-Cabrera et al., 1997; Lopez et al., 1997). Numerical solutions of the Fick's law differential diffusion equation with constant (Daud et al., 1997) or moisture and temperature dependant (Zogzas and Maroulis, 1996) diffusivity also have been used for the moisture diffusivity estimation.

The main problem in the moisture diffusivity determination by classical or inverse methods is the difficulty of moisture content measurements. Local moisture content measurements are practically unfeasible especially for small drying objects. Standard drying curves measurements (body mean moisture content during the drying) are complex and of low accuracy. Instead, accurate and easy to perform single thermocouple temperature measurement can be used. The main idea of the present method is to take advantage of the relation between the heat and mass (moisture) transport processes within the drying body and from its surface to the surroundings. Then, the moisture diffusivity estimation can be performed on the basis of a temperature response by using an inverse approach. Kanevce, Kanevce and Dulikravich (2000) and Dantas et al. (1999) recently analyzed this idea of the moisture diffusivity estimation by temperature response of a drying body.

The objective of this paper is 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. In order to realize this analysis the sensitivity coefficients and the sensitivity matrix determinant have been calculated for the characteristic drying regimes and drying body dimensions.

In this paper, the solution of the inverse problem of estimating the moisture content and temperature-dependent moisture diffusivity is presented. The drying process considered here is described by the non-linear one-dimensional Luikov's equations. The associated direct problem is solved numerically. 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. As a representative drying body, a mixture of bentonite and quartz sand with known thermophysical properties has been chosen.

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MATHEMATICAL MODEL OF DRYING: DIRECT PROBLEM

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

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

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

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

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

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

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

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

$$j_q = h(T_a - T_{x=L})$$
$$j_m = h_D(C_{x=L} - C_a) \qquad (5)$$

The water vapor concentration in the drying air, C_a is calculated by

$$C_a = \varphi \cdot p_s(T_a)/461.9/(T_a + 273) \qquad (6)$$

and, the water vapor concentration of the air in equilibrium with the free surface of the body is calculated by

$$C_{x=L} = a(T_{x=L}, X_{x=L}) \cdot p_s(T_{x=L})/461.9/(T_{x=L} + 273) \qquad (7)$$

The water activity, a, or the equilibrium relative humidity of the air in contact with the surface at temperature $T_{x=L}$ and moisture content $X_{x=L}$ is calculated from experimental water sorption isotherms.

The problem is symmetrical, and boundary conditions on the mid-plane of the plate ($x = 0$) are

$$\left(\frac{\partial T}{\partial x}\right)_{x=0} = 0, \quad \left(\frac{\partial X}{\partial x}\right)_{x=0} = 0 \qquad (8)$$

In order to approximate the solution of Eqs. (1-2), an explicit procedure has been used (Kanevce, Kanevce and Dulikravich, 2000). The nonlinear term has been expanded to

$$\frac{\partial}{\partial x}\left(D\frac{\partial X}{\partial x}\right) = D\frac{\partial^2 X}{\partial x^2} + \frac{\partial D}{\partial x}\frac{\partial X}{\partial x} \qquad (9)$$

The derivatives with respect to time have been represented using forward differencing at the grid-point (i,j). All the first- and second-order space derivatives have been approximated at time level (j) using central differencing. The moisture diffusivity, D, in the first term of Eq. (9) has been assigned its value at the grid point (i,j). Central differencing has been also applied to the boundary conditions space derivatives.

MOISTURE DIFFUSIVITY ESTIMATION: INVERSE PROBLEM

The estimation methodology used is based on minimization of the ordinary least square norm

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

Here, $Y^T = [Y_1, Y_2, \ldots, Y_{imax}]$ is the vector of measured temperatures and $T^T = [T_1(P), T_2(P), \ldots T_{imax}(P)]$ is the vector of estimated temperatures at time t_i (i = 1, 2, ..., imax), while $P^T = [P_1, P_2, \ldots P_N]$ is the vector of unknown parameters, imax is the total number of measurements, and N is the total number of unknown parameters (imax ≥ 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 and has been applied to a variety of inverse problems (Mejias, Orlande and Ozisik, 1999). It belongs to a general class of damped least square methods (Beck and Arnold, 1977). The solution for vector P is achieved using the following iterative procedure

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

$$J = \begin{bmatrix} \frac{\partial T_1}{\partial P_1} & \cdots & \frac{\partial T_1}{\partial P_N} \\ \cdot & & \\ \frac{\partial T_{i\,max}}{\partial P_1} & \cdots & \frac{\partial T_{i\,max}}{\partial P_N} \end{bmatrix} \qquad (12)$$

Near the initial guess, the problem is generally ill-conditioned and large damping parameter is chosen thus making term μI large as compared to term $J^T J$. The term μI damps instabilities due to ill-conditioned character of the problem. 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 (Beck and Arnold, 1977; Mejias, Orlande and Ozisik, 1999). In fact, this method compromises between the steepest descent and Gauss method by choosing μ so as to follow the Gauss method

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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 (Pfafl and Mitchel, 1969).

RESULTS AND DISCUSSION

For the direct problem solution, the system of Eqs. (1) and (2) with the initial conditions, Eq. (3), and the boundary conditions, Eqs. (4) and (8), has been solved numerically for a model material, a mixture of bentonite and quartz sand with the known, experimentally determined (Kanevce et al., 1980) thermophysical properties. 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 thermo-diffusion is small and can be ignored. It was also concluded that the Luikov's system of two simultaneous partial differential equations could be used. In this case, the transport coefficients can be treated as constants except for the moisture diffusivity. The appropriate mean values for the model material are

$\rho_s = 1738\ kg/m^3$

$c = 1550\ J/K/kg\ db$

$k = 2.06\ W/m/K$

$\Delta H = 2.31 \cdot 10^6\ J/kg$

$\varepsilon = 0.5$

$\delta = 0.$

The following empirical expression can describe the experimentally obtained relationship for the moisture diffusivity (Kanevce et al., 1980)

$$D = 9.0 \cdot 10^{-12} \cdot X^{-2} \cdot \left(\frac{T + 273}{303} \right)^{10} \quad (13)$$

The experimentally obtained desorption isotherms of the model material (Kanevce, 1981) is presented by the empirical equation

$$a = 1 - exp(-1.5 \cdot 10^6 \cdot (T + 273)^{-0.91} \cdot \quad (14)$$

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 uncertainty in the estimated moisture diffusivity is smaller than the uncertainty in the mass transfer coefficient. Besides, the moisture content of the surface of the body is practically constant, near equilibrium moisture content very short after the beginning of the drying (Fig. 4). In conclusion, constant heat and mass transfer coefficients are taken for the purpose 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 this paper the moisture diffusivity of the model material has been represented by the following function of temperature and moisture content

$$D = \frac{D_1}{D_2 + X^2} \cdot \left(\frac{T + 273}{303} \right)^{10} \quad (15)$$

where D_1 and D_2 are constants. For the inverse problem investigated here, values of D_1 and D_2 are regarded as unknown and all other quantities appearing in direct problem formulation are assumed to be known. Thus, $P^T = [D_1, D_2]$.

In order to investigate influence of the boundary conditions, determinant of the sensitivity matrix J^TJ with normalized elements was calculated.

$$[J^TJ]_{m,n} = \sum_{i=1}^{i\,max} \left(P_m \frac{\partial T_i}{\partial P_m} \right) \left(P_n \frac{\partial T_i}{\partial P_n} \right), \ m, n = 1,N \quad (16)$$

The sensitivity coefficients analysis has been carried out for the plate of thickness $2L$, with initial moisture content of $X(x, 0) = 0.20\ kg/kg$ and initial temperature $T(x,0) = 20.0\ ^oC$. In order to investigate the influence of the boundary conditions, the drying air bulk temperature, T_a and velocity, V_a, have been varied (Table 1.). The relative humidity of the drying air was $\varphi = 0.12$.

Table 1 Drying air conditions

where the water activity, a, represent the relative humidity of the air in equilibrium with the drying object at temperature T and moisture content X.

Any appropriate relations for the convective heat and mass transfer coefficients (h and h_D) can be used. An infinite or constant mass transfer coefficient has been mostly used in the moisture diffusivity estimation based on the experimental drying curves relation to the solution of the differential diffusion equation. The presented method allows a moisture and temperature dependent mass transfer coefficient to be used. The best approach should be if the appropriate relations are obtained by comparison of the known with individual experiments

3 mm	6 mm	$T_a[^0C]$	V_a[m/s]	$h[W/m^2K]$	$h_D 10^2[m/s]$
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 depicts the mean convective heat and mass transfer coefficients (h and h_D) obtained for an 80 *mm* long plate (Luikov, 1972). The test cases have been repeated for two different thicknesses of the drying body, $2L = 3.0$ and 6.0 *mm*.

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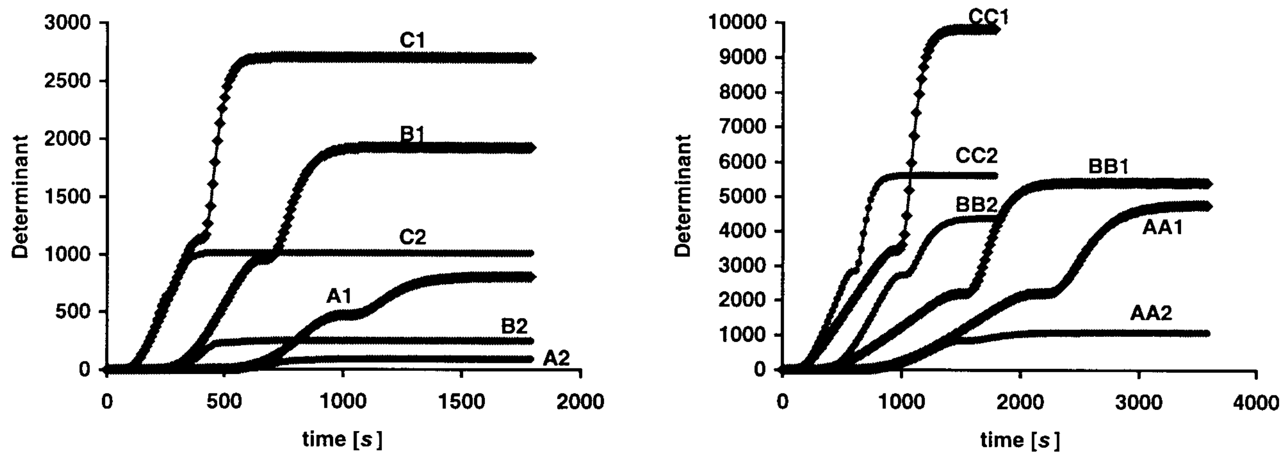

Figure 1. Sensitivity determinants

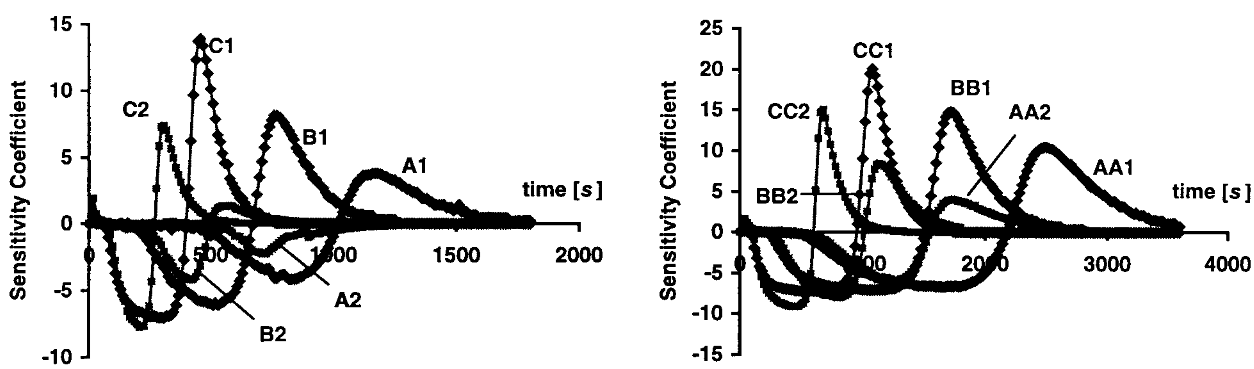

Figure 2. Sensitivity coefficients

Figure 1 shows the determinant of the sensitivity matrix J^TJ with normalized elements and Fig. 2 the relative sensitivity coefficient $D_l \partial T_i / \partial D_l$, i = 1, 2,..., imax, 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, 2000) when the body moisture content is nearly equal to the equilibrium. After that, small evaporation rate and fast drying rate decrease, while fast body temperature increase occurs. For the drying times shorter than that, the problem is ill-posed and local minimum can be obtained depending on the initial guesses. The second plateau corresponds to the moment when nearly equilibrium temperature has been obtained.

The tendency of increasing determinant as well as sensitivity coefficients with increase in drying air velocity (for example, cases A1, B1, and C1) is obvious. This can be explained by the increase of mass transfer Biot number ($Bi_D = h_D L/D$) and the moisture content gradients inside the body. The heat transfer Biot number ($Bi = hL/k$) and the temperature gradient are very small in all the cases. The heat transfer Biot number was of the order 0.02 to 0.12 depending on the boundary conditions and the plate thickness. The mass transfer Biot number ranged from 60 to 2 x 10^6. It changes with local moisture content and temperature during the drying from 3 x 10^5 to 60 for the case A1 and from 9 x 10^5 to 180 for the case C1. Under these conditions, mainly the moisture diffusivity and dimensions of the body govern the process of drying.

The influence of the drying air temperature also can be explained through the mass transfer Biot number and the

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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 coefficients with increasing drying air temperature (Cases C1 and C2). On the other hand, higher drying air temperature leads to a shorter drying time and consequently smaller computational time for a direct problem. The same conclusion remains when plate thickness increases (Cases C1 and CC1).

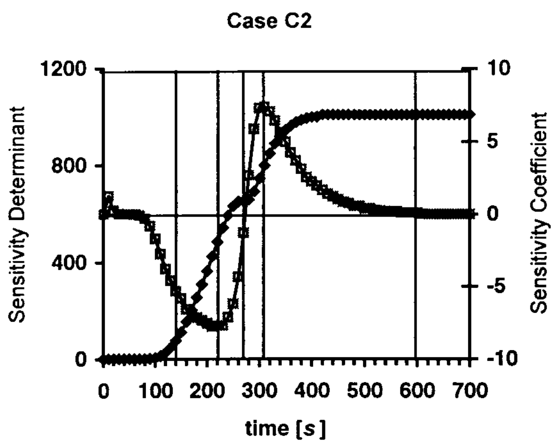

Figure 3. Sensitivity coefficient (square symbols) and determinant (diamond symbols) for Case C2

For the estimation of the moisture diffusivity available, the transient readings of the temperature sensor located in the mid-plane have been considered. The simulated experimental data ware obtained from the numerical solution of the direct problem presented above, 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 ^oC was added to the numerical temperature response.

In this paper, parameters D_1 and D_2 have been estimated using the test case having the shortest drying time (C2). Five drying times, corresponding to Fig. 3 and Fig. 4 have been considered: the end of drying and the maximum determinant value (600 seconds), the maximum sensitivity coefficient value (310 seconds), the first determinant plateau value and zero sensitivity coefficient value (272 seconds), the maximum negative sensitivity coefficient value (220 seconds), and the drying time below that (140 seconds).

The temperature response had constant time-step of two seconds. Thus, different numbers of temperature measurements have been taken for the cases with different drying times. The number of the space grid points was 41 in all the drying process calculation schemes.

The initial guess for all the cases was the same ($D_{1init} = 5.0\cdot10^{-11}$ and $D_{2init} = 0.1$). It was far from the exact values of parameters.

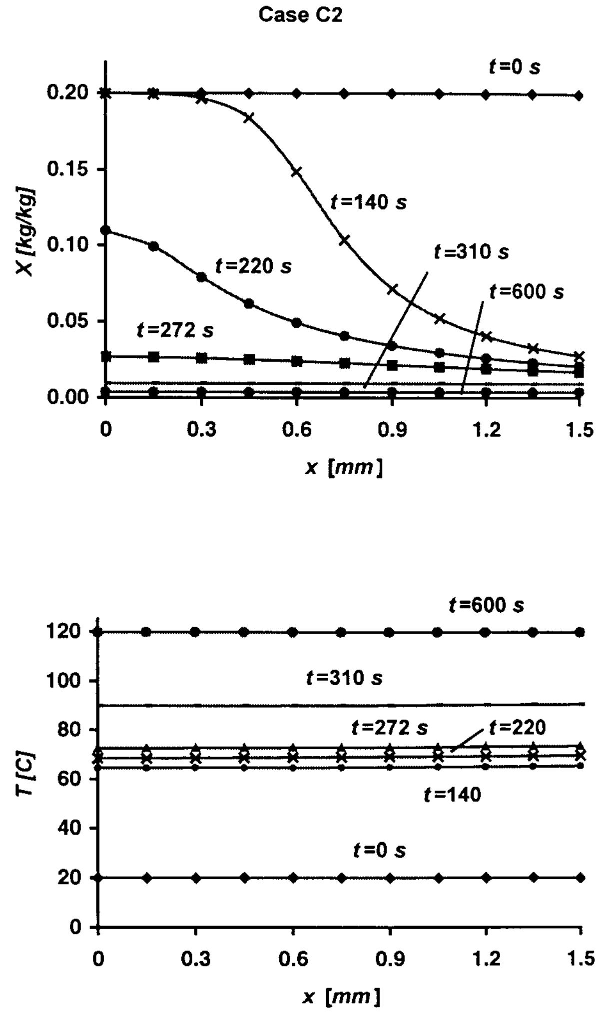

Figure 4. Transient moisture content and temperature profiles

Table 2 shows the computationally obtained results. For comparison, the values of exact parameters and the values estimated with "exact" (without noise) temperature data are also shown. The results show the tendency of increasing the accuracy of computing D_1 with the increase in the value of the determinant. The relative error has been calculated as $\varepsilon_1(\%) = 100\cdot|(D_1-D_{1ex.})/D_{1ex}|$. The accuracy of computing D_2 depends on the X^2 value. It changes from the beginning to the end of drying, $\varepsilon_2 = ((X^2+D_2)-X^2)/X^2 = D_2/X^2$.

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Table 2. Estimated parameters (Case C2)

	$t[s]$	$D_1.10^{12}$	$\varepsilon_1[\%]$	D_2	$\varepsilon_2[\%]$	RMS
Exact values		9.000		0.0		
Errorless data	272	9.003	0.03	$4.0 \cdot 10^{-7}$	0.004	0.00059
Data with error (σ=0.5)	140	10.355	15.06	$3.2 \cdot 10^{-4}$	3.2	0.44353
	220	8.799	2.23	$1.5 \cdot 10^{-1}$	1500.	1.7139
	272	8.978	0.24	$3.6 \cdot 10^{-7}$	0.004	0.49126
	310	9.015	0.17	$2.9 \cdot 10^{-7}$	0.003	0.50813
	600	9.012	0.13	$8.3 \cdot 10^{-7}$	0.008	0.49728

The mean relative error of D_2 has been calculated with the mean moisture content value X_m, as $\varepsilon_2(\%) = 100 \cdot | D_2/X_m^2 |$. As a mean moisture content value during the drying, $X_m = 0.10$ *kg/kg* has been utilized for all the cases. The mean accuracy of the computed D_2 is very high for all the well-posed cases.

It can be seen that very good agreement is achieved for all the cases with drying time equal or longer than that corresponding to the first plateau on the determinant curve. For the drying times shorter than that, the problem is ill-posed and a local minimum is often obtained.

CONCLUSIONS

The method for estimation of moisture content and temperature-dependent moisture diffusivity that is based on thermal transient response of a drying body was presented. It uses an inverse approach that utilizes the Levenberg-Marquardt method for evaluation of unknown parameters in the moisture diffusivity dependence on moisture and temperature. The results of the numerical experiments show good agreement between evaluated and exact parameter values and confirm the validity of the proposed method.

The tendency towards increasing the determinant as well as sensitivity coefficient with drying air velocity and drying body thickness increasing and with drying air temperature decreasing was obtained. It was 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 are very small ($Bi \leq 0.12$) in all the cases. The mass transfer Biot number ranged from 60 to 2 x 10^6. Under these conditions it is the moisture diffusivity and dimensions of the body that govern the process of drying.

The influence of the drying air velocity increase on the value of the sensitivity determinant and drying time leads to the computational time decrease. The opposite influence of the drying air temperature and drying body thickness on the determinant and drying time suggests that more work needs to be done in defining their optimal values concerning the minimization of the computing time.

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