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## Modeling of the primary microwave freeze-drying of granular solids using a fixed grid approach

### Introduction

Freeze-drying is a sophisticated method of dehydration used to obtain the highest quality preserved biological materials, pharmaceuticals and foodstuffs. It is carried out in three stages: pre-freezing, sublimation drying under vacuum (primary drying), and residual water desorption (secondary drying).

Application of microwave energy in the process accelerates considerably the rate of drying. However, it carries also the risk of corona discharge and non-uniform heating.

The modeling of the microwave freeze-drying requires taking into account both energy and mass transfer, volumetric generation of heat source in the sample as well as the existence of the moving boundary (sublimation ice front) – the interface separating the dried and the frozen material regions.

In numerical modeling of the process, the moving boundary problem must be solved. The two fixed grid approaches are proposed in this paper: the variable time step method and *Landau's* transformation. The simulations are performed for Sorbonorit 4 activated carbon, which is a representative of a granular dielectric containing internal porosity.

### Mathematical modeling

The one-dimensional mathematical model of coupled heat and mass transfer in microwave freeze-dried material is developed [1] (Fig. 1).

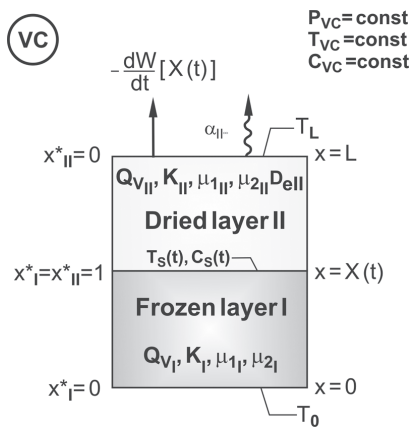


Fig. 1. Physical model of the primary microwave freeze-drying

The following assumptions are made to simplify the model:

- initial arbitrary position of moving boundary  $X_{ini}$  is assumed;
- ice front retreats uniformly during the process as a result of sublimation and diffusion of water vapors from the interface toward exposed surface;
- in the frozen layer I heat is transferred by conduction whereas both conduction and convection take place in the dried layer II;
- the electric field strength in the dried material is uniform in the whole sample.

### Governing equations

Heat transfer in the frozen and dried regions of the material is governed by:

$$\frac{\partial T_i}{\partial t} = a_{ei} \frac{\partial^2 T_i}{\partial x^2} + \frac{Q_{vi}}{\rho_{ei} c_{pei}} \quad (1)$$

$$\frac{\partial T_{II}}{\partial t} = a_{eII} \frac{\partial^2 T_{II}}{\partial x^2} - \frac{c_{ps} N_w}{\rho_{buII} c_{peII}} \frac{\partial(T_{II})}{\partial x} + \frac{Q_{vII}}{\rho_{buII} c_{peII}}, \quad (2)$$

where steady capacity of internal heat source  $Q_{vi}$  is defined as [2]:

$$Q_{vi} = K_i(T_i)E^2 \text{ for } i = I, II. \quad (3)$$

Dissipation coefficient  $K_i(T_i)$  is simplified by linear relation:

$$K_i(T_i) = \pi f \epsilon_0 \epsilon_i''(T_i) \approx \mu_1 T_{II} + \mu_2, \quad (4)$$

where coefficients  $\mu_1$  and  $\mu_2$  are determined experimentally on the basis of calorimetric procedure [3].

In the dried region, diffusion of water vapor is governed by:

$$D_{eII} \frac{\partial^2 C}{\partial x^2} = \frac{\partial C}{\partial t} \quad (5)$$

The mass flux density follows *Fick's* law, and the effective diffusivity  $D_{eII}$  is combination of the *Knudsen* and molecular diffusivity [4].

The energy and mass balance at the sublimation front can be expressed as:

$$q_s(t) = -k_{ei} \left( \frac{\partial T_i}{\partial x} \right)_{x=X} + k_{eII} \left( \frac{\partial T_{II}}{\partial x} \right)_{x=X} = N_w(t) \Delta h_s \quad (6)$$

The displacement of the interface position is depended on the rate of sublimation:

$$N_w(t) = -(W_{mi} - W_{eq}) \rho_{buII} \frac{dX(t)}{dt} \quad (7)$$

### Initial and boundary conditions

The formulated model is solved together with the following initial conditions:

- initial moving boundary position:

$$X(t) = L - \delta \quad (8)$$

- initial temperature profiles:

$$T_i(x,0) = T_{mi}, \text{ for } 0 \leq x \leq X \quad (9)$$

$$\frac{T_L(0) - T_{II}(x,0)}{T_L(0) - T_{mi}(0)} = \frac{L - x(0)}{\delta} \text{ for } X \leq x \leq L \quad (10)$$

- initial concentration profile:

$$C = C_{vc} - \left( \frac{\partial C}{\partial x} \right)_{x=X} (L - x) \text{ for } X \leq x \leq L \quad (11)$$

The following boundary conditions are assumed:

- at the bottom boundary of the material:

$$\left( -k_{ei} \frac{\partial T_0}{\partial x} \right)_{x=0} = 0 \quad (12)$$

- at the exposed material surface:

$$k_{eII} \left( \frac{\partial T_{II}}{\partial x} \right)_{x=L} = \alpha_{I\infty} (T_{vc} - T_L) \quad (13)$$

$$C_L(t) = C_{vc} \quad (14)$$

- at the moving front:

$$C_s(t) = f(T_s) \quad (15)$$

**Methods of model solution**

The formulated mathematical model of the microwave freeze-drying was solved numerically for process parameters and material properties collected in tab. 1.

Tab. 1. Thermophysical properties of the drying system

Material thickness $L$	0.01 m	Initial ice front position $\delta$	0.0097 m
Elec. field strength $E$	1000 V/m	Total pressure $P$	100 Pa
Ambient temp. $T_{VC}$	20°C	Initial temperature $T_{ini}$	-20°C
Heat trans. coeff. $\alpha_{H\infty}$	20 W/(m <sup>2</sup> K)	Bed porosity $\epsilon$	0.71 m <sup>3</sup> /m <sup>3</sup>
Avg. moist. cont. $W_{eq}$	0.05 kg/kg	Eq. moisture content $W_{ini}$	1.63 kg/kg
Layer parameter	Frozen layer $i = I$	Dried layer $i = II$	
Thermal diffusivity $a_{ei}$	1.54 m <sup>2</sup> /s (100 Pa)	0.30 m <sup>2</sup> /s (100 Pa)	
Thermal conductivity $k_{ei}$	1.60 W/mK (100 Pa)	0.094 W/mK (100 Pa)	
Parameter in Eq. (4) $\mu_{1i}$	0.00090 W/(KmV <sup>2</sup> )	0.00079 W/(KmV <sup>2</sup> )	
Parameter in Eq. (4) $\mu_{2i}$	0.41148 W/(mV <sup>2</sup> )	0.30905 W/(mV <sup>2</sup> )	
Density $\rho_{ei}$	1048 kg/m <sup>3</sup>	400 kg/m <sup>3</sup>	

**Variable Time Step (VTS) method**

The variable time step method is a spatial fixed grid approach used in this work together with the implicit finite-difference *Crank-Nicolson* scheme. Utilization of this method means, that in each step of computations the sublimation ice front should move exactly one grid space from the previous location. In order to determine the adequate time step corresponding to this movement, the following simultaneous heat and mass balance at the moving boundary must be fulfilled:

$$-k_{ei} \frac{T_{xj} - T_{I_{x+1,j}}}{\Delta x} - k_{eII} \frac{T_{xj} - T_{II_{x+1,j}}}{\Delta x} + (W_{ini} - W_{eq})\rho_{huu}\Delta h_s \frac{\Delta X}{\Delta t} - \rho_{ei}c_{peI} \frac{\Delta X}{\Delta t} (T_{xj} - T_{I_{x+1,j}}) = 0 \quad (16)$$

The individual terms in Eq. (16) correspond to heat flux densities transferred from the frozen and dried material regions, heat flux utilized by sublimation and heat required to raise the temperature of interface node from previous time step to current one.

Since the finite difference schemes of Eqs. (1)-(2), (5)-(7) together with the initial and boundary conditions (8)-(15) constitute the linear equations set, it can be easily solved by the tridiagonal algorithm. In each calculation step, value of the actual time step  $\Delta t$  is determined iteratively by bisection method as a root of Eq. (16).

The simulations results of microwave freeze-drying of *Sorbonorit 4* using the VTS approach are shown in fig. 2.

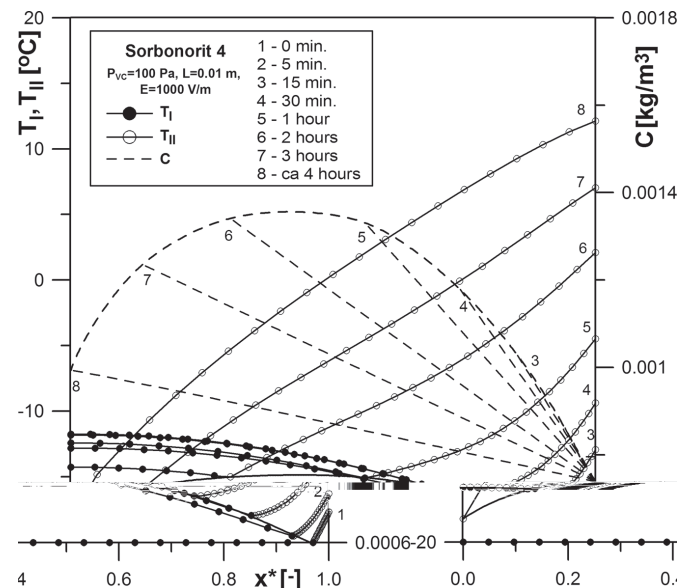


Fig. 2. Material temperatures and water vapor concentrations vs. dimensionless position coordinates in selected process times – VTS method

**Landau's transformation method**

Landau's transformation immobilizes mathematically position of the sublimation front by the following definitions of dimensionless position coordinates in the frozen and dried region:

$$x_i^* = \frac{x}{X(t)}, \quad x_{ii}^* = \frac{L-x}{L-X(t)} \quad (17)$$

Substitution of above definitions into model equations results in complex relations which, after the discretization by *Crank-Nicolson* scheme, become nonlinear and must be solved iteratively. In this paper *Mathcad* software was used to overcome calculation difficulties. Obtained typical temperature and water concentration profiles in material are presented in fig. 3.

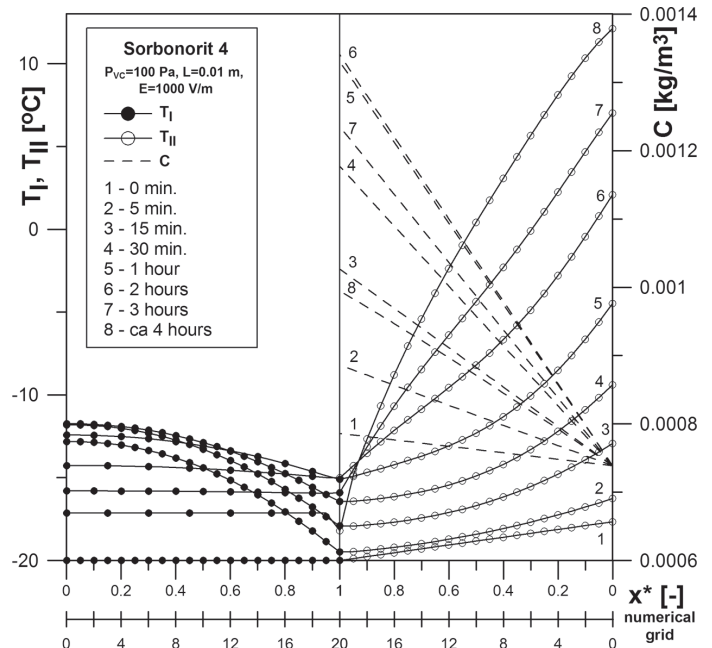


Fig. 3. Material temperatures and water vapor concentrations vs. dimensionless position coordinates in selected process times – Landau's transformation

Small number of numerical nodes, i.e. 20 per region was sufficient to obtain results comparable with those in the VTS approach. However, calculations lasted much longer and it can be shortened by increasing the time step periodically [5].

**Conclusions**

In the modeling of the microwave freeze-drying, moving boundary problem must be solved. Fixed grid approaches are used to simplify numerical solutions. Landau's transformation immobilizes ice front position but results in mathematically complex form which is difficult to solve. The variable time step is an alternative approach which seems to be the most effective method. One-dimensional model is accurate enough to describe the microwave freeze-drying of slabs when temperature dependent dielectric properties of material are taken into account.

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