



## On the computer simulation of heat and mass transfer in vacuum freeze–drying

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**Abstract.** The paper is devoted to studying the problem of freeze–drying which is a process of the dehydrating frozen materials by sublimation under high vacuum. The mathematical and the computer models describing this process are presented. The discretizations used and the numerical treatment of the corresponding ordinary and partial differential equations is discussed. The results of some test experiments and the corresponding analysis can be found.

**Key words:** freeze–drying, heat and mass transfer, ordinary and partial differential equations, Runge-Kutta methods, heat conduction equation, finite element and finite difference methods

### 1 Introduction

Freeze–drying is a process by which a solvent is removed from a frozen material by sublimation under high vacuum ([4, 8]). The process of dehydration under vacuum starts from a frozen state. The heat is supplied to the drying material by: (i) conduction, (ii) radiation or (iii) both conduction and radiation but at a low rate in order to avoid the local melting. The main advantages of such kind of drying are:

- higher food quality of the dry product due to the minimum loss of flavor and aroma;
- the chances of the microbial to growth are minimized due to the water absence;
- no thermal and oxidizing processes in the dried product, etc.

In general, two different containers (cameras) are needed for the process of freeze–drying. The first one (food container) is for the product which will be dried. The second container consists of two subareas. One of them is fill–in with some material for the sorbtion of the water molecules, which come from the food container, and another is vacuum. The following three main stages can be seen in the process of freeze–drying:

1. Activation (dehydration) of the sorbent and preparation of the product to be dried. During this phase:
  - the freezing (drying) container is filled with the product to be dried and vacuumed;
  - the adsorbent which is located in the second container is warmed up, vacuumed and cooled to a room temperature.
2. Self-freezing of the product to be dried.
3. Drying in the conditions of an uniform sublimation of water steam from the product and their deposition in the sorbent.

This paper is devoted to study numerically (i) the *drying front movement in the freeze dryer*, (ii) *the temperature fields in the adsorbent camera* and (iii) *the transfer of the sublimated water molecules and their retention in the sorbent granules*.

In this study *Zeolites granules* are used to sublimate the water molecules. Zeolites are a special type of silica-containing material which have a porous structure that makes them valuable as adsorbents and catalysts (for more details see e. g. [10]). They are used in different environmental problems, e. g. for reducing methyl bromide emissions, cleaning water, etc. Let us mention, that in practice not only natural zeolites but also synthetic ones can be used in the process of freeze-drying. Both, natural and synthetic zeolites have a rigid, three-dimensional crystalline structure (similar to honeycomb) consisting of a network of interconnected tunnels and cages ([11]).

The mathematical model of the process of freeze-drying is described by a system of time-dependent differential equations. The model has a hierarchical structure and a splitting procedure according to the technological processes involved, is applied.

The rest of the paper is organized as follows. In Section 2 the mathematical model is discussed. The discretization used and the numerical treatment of the mathematical model is presented in Section 3. The results of a test experiment are given in Section 4. Finally, some conclusions and plans for future work can be found in Section 5.

## 2 The mathematical model

As was mentioned above, the process of freeze-drying has a hierarchical structure. Therefore, a splitting procedure according to the technological subprocesses involved, is applied.

### 2.1 Modelling the drying front in the freeze camera

The process of freeze-drying starts with the *self-freezing* of the product which will be dried. This first step of the process is relatively very fast and it takes short time after the connection between the *freezing* and *adsorption* cameras is open. There are three possibilities of heating the frozen product after this initial

stage (see Fig. 1). One can use a *radiation heater* (see Fig. 1, the left picture). In this case (so called *radiation heating*) the heating is coming from above and the dried layer is starting from above. If for heating is used a plate from bellow, then two different cases can occur. In the first one (see Fig. 1, the picture in the middle), the dried layer is starting from bellow and in the second case (see Fig. 1, the right picture) the dried layer is starting from above. Let us denote with  $t$  the time after starting of the process of drying, with  $L$  – the height of the frozen product and with  $Y = Y(t)$  the thickness of the dried layer at time  $t$ .

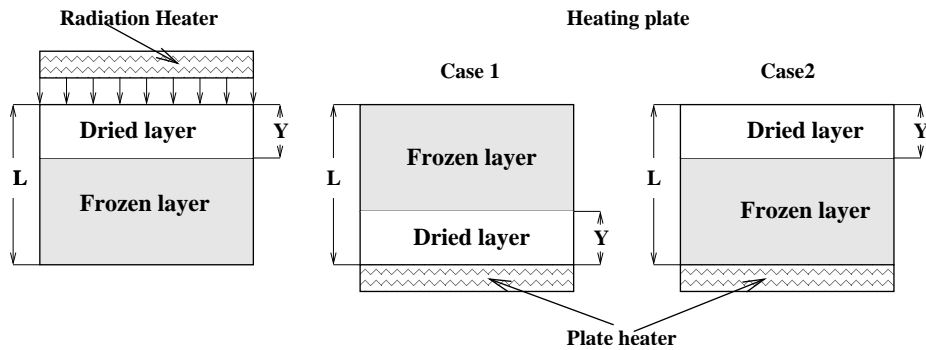


Fig. 1. Drying cases

Then, the drying front in the freezing camera can be described with one of the following three ordinary differential equations (ODE):

– radiation heating:

$$\frac{T_r - T_{sub}}{\frac{1}{h_r} + \frac{Y}{k_{cr}}} = \lambda \frac{P_{sub} - P_c}{\frac{1}{k_m} + \frac{Y}{\beta_{cr}}} = \rho \varepsilon \frac{dY}{dt}; \tag{1}$$

– heating plate, case 1:

$$\frac{T_p - T_{sub}}{\frac{1}{h_c} + \frac{Y}{k_{cr}}} = \rho \varepsilon \frac{dY}{dt}; \tag{2}$$

– heating plate, case 2:

$$\frac{T_p - T_{sub}}{\frac{1}{h_c} + \frac{L-Y}{k_{ice}}} = \lambda \frac{P_{sub} - P_c}{\frac{1}{k_m} + \frac{Y}{\beta_{cr}}} = \rho \varepsilon \frac{dY}{dt}. \tag{3}$$

The following notations are used in the last three equations:

- $T_r$  – radiation temperature;
- $T_p$  – plate temperature;
- $T_{sub}$  – sublimation temperature;
- $P_{sub}$  – water vapour pressure at the sublimation temperature;

- $P_c$  – water vapour pressure at the condensation temperature;
- $h_r$  – radiation heat transfer coefficient;
- $h_c$  – contact heat transfer coefficient;
- $k_{cr}$  – thermal conductivity of the dried layer;
- $k_{ice}$  – thermal conductivity of the frozen layer;
- $\rho$  – density coefficient;
- $\varepsilon$  – moisture content;
- $\lambda$  – latent of sublimation of the ice;
- $\beta_{cr}$  – permeability of the dried layer;
- $k_m$  – external mass transfer coefficient.

## 2.2 Modelling the process of heat and mass transfer

The process of heat and mass transfer in the adsorbent camera is described by the nonlinear partial differential equation (PDE) of parabolic type:

$$c\rho\frac{\partial T}{\partial t} = \mathcal{L}T + f(x, t, T), \quad x \in \Omega, \quad t > 0, \quad (4)$$

where

$$\mathcal{L}T = \sum_{i=1}^d \frac{\partial}{\partial x_i} \left( k(x, t) \frac{\partial T}{\partial x_i} \right). \quad (5)$$

The following notations are used in (4) and (5):

- $T(x, t)$  – unknown distribution of the temperature;
- $d$  – dimension of the space ( $d = 2$  in this study);
- $\Omega \in R^d$  – computational domain (see Fig. 3);
- $k = k(x, t) > 0$  – heat conductivity;
- $c = c(x, t) > 0$  – heat capacity;
- $\rho > 0$  – material density;
- $f(x, t, T)$  – right-hand side function, which is responsible for the non-linear process of transfer of water molecules in the adsorption container. This function couples the two systems of equation (one of (1), (2) or (3), and (4)).

Let us mention, that the adsorbent camera consists of several subparts, and each of them has their own conductivity, capacity and density coefficients.

*Initial* (6) and *boundary* (7) conditions are assigned to the parabolic equation (4):

$$T(x, 0) = T_0(x), \quad x \in \Omega, \quad (6)$$

$$T(x, t) = \mu(x, t), \quad x \in \Gamma \equiv \partial\Omega, \quad t > 0, \quad (7)$$

where  $T_0(x)$  is the initial distribution of the temperature in the computational domain and  $\mu(x, t)$  is most often the surrounding of the freeze-drying device temperature.

### 3 Numerical treatment

#### 3.1 Numerical treatment of the drying front equation

Let us write the three cases in describing the drying front in the freezing camera presented by (1), (2) or (3) in the form:

$$\frac{dY}{dt} = f(t, Y), \quad t \in [0, T], \quad (8)$$

where the right-hand-side function  $f$  is one of the left-side-functions in (1), (2) or (3) depending on the case of drying, dividing by  $\rho \varepsilon \lambda$ . An uniform discretization of the time interval  $t_i = t_0 + i\tau$ , where  $\tau$  is the time-step, is used. For the numerical solution of (8) the Runge-Kutta method of order fourth is applied ([6]). Then

$$Y_{n+1} = Y_n + \sum_{r=1}^4 p_r k_r, \quad (9)$$

where  $k_j = \tau f\left(t_n + \alpha_j \tau, Y_n + \sum_{l=1}^{j-1} \beta_{j,l} k_l\right)$ ,  $j = 1, 2, 3, 4$ , and the parameters in the last formulas are taken according to the so-called *3/8-Rule* as follows:

$$p_1 = p_4 = \frac{1}{8}, \quad p_2 = p_3 = \frac{3}{8}, \quad \alpha_1 = 0, \quad \alpha_2 = \frac{1}{3}, \quad \alpha_3 = \frac{2}{3}, \quad \alpha_4 = 1,$$

$$\beta_{2,1} = \frac{1}{3}, \quad \beta_{3,1} = -\frac{1}{3}, \quad \beta_{3,2} = \beta_{4,1} = \beta_{4,3} = 1, \quad \beta_{4,2} = -1.$$

#### 3.2 Numerical treatment of the heat and mass transfer front equation

For the numerical solution of the above discussed boundary value problem the Finite Element Method (FEM) in space is used ([2]). In fact, the Courant linear finite elements are chosen in this study. After the space discretization, the time derivatives are discretized via finite differences and well known Crank-Nicolson scheme is used ([7]). The computational domain is discretised via triangle finite elements (see e.g. Fig. 4) using the computer mesh generator **Triangle** ([3, 9]). Options which are used for the mesh generating (generation of the main grid) are: *minimal angle of a triangle* (the most often used value is  $30^\circ$ ) and *maximal area of a single triangle* (different for the different subdomains and depending of the geometry). During the single run this basic grid can be refined depending on a run parameter with a factor of  $k = 1, 2, 3, \dots$  and one can get respectively from each basic triangle 4, 16, 64, ... finer triangle elements, after bisection of the basic grid triangles sides.

Let us denote with  $K$  and  $M$  the stiffness and mass matrices coming from the Finite Element approach. In our case they can be written in the forms:

$$K = [K_{ij}]_{i,j=1}^N = \left[ \int_{\Omega} k \nabla \Phi_i \cdot \nabla \Phi_j dx \right]_{i,j=1}^N,$$

$$M = [M_{ij}]_{i,j=1}^N = \left[ \int_{\Omega} c\rho \Phi_i \Phi_j dx \right]_{i,j=1}^N.$$

Then, the parabolic equation (4) can be written in matrix form as:

$$M \frac{dT}{dt} + KT = F, \quad (10)$$

where

$$F = \left[ \int_{\Omega} f(x, t, T) \Phi_j dx \right]_{j=1}^N$$

If we denote with  $\tau$  the time step, with  $T^{n+1}$  the solution (temperature) on the current time level and with  $T^n$  the solution on the previous time level, and do an approximation of the time derivative in (10) we will obtain the following system of linear algebraic equations for the nodal values of  $T^{n+1}$ :

$$\left( M + \frac{K\tau}{2} \right) T^{n+1} = \left( M - \frac{K\tau}{2} \right) T^n + \tau \frac{F^{n+1} + F^n}{2}. \quad (11)$$

The Preconditioned Conjugate Gradient (PCG) method with a Modified Incomplete Cholesky (MIC(0)) Preconditioner ([1, 5]) is used to solve the system of algebraic equations (11).

### 3.3 Structure of the computer model

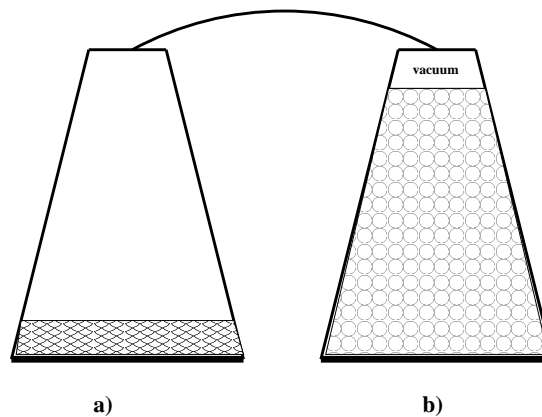
The computer code realizing the above presented algorithm consists of two main modules according to the splitting done. The structure of the code is as follows:

- Numerical solution of the drying front equations in drying freeze camera (Runge–Kutta methods for solution of nonlinear ODE);
- Numerical solution of the heat–mass transfer equation in adsorbent camera (Crank–Nikolson method and FEM for parabolic PDEs):
  - triangulation of the computational domain (computer mesh generator **Triangle**);
  - generation of the element stiffness and mass matrices and vectors;
  - assembling of the global stiffness and mass matrices and vectors;
  - solution of the systems of linear algebraic equations (PCG method with MIC(0) preconditioner);
  - visualization of the output results.

The program modules are developed using the algorithmic languages *Fortran* and  $C^{++}$  under *Linux/Unix* operating system.

#### 4 Numerical test on a real-life experiment

Many numerical experiments with known exact solutions were run in order to fix the mesh and time parameters of the computer model. For the final test of the numerical algorithm discussed above, an operating experimental unit was used (see Fig. 2). In this experiment the case where for the heating is used a plate from bellow and the dried layer is started from bellow is used (see Fig. 1, case in the middle of the picture). The product which is subject to drying is put in the left container. In the case study this container is a *glass flask* and the drying product is *grated carrots* (see Fig. 2a)). The right container is the absorption camera (in this study it is again made from *glass*), where the zeolite granules are placed (see Fig.2b)).



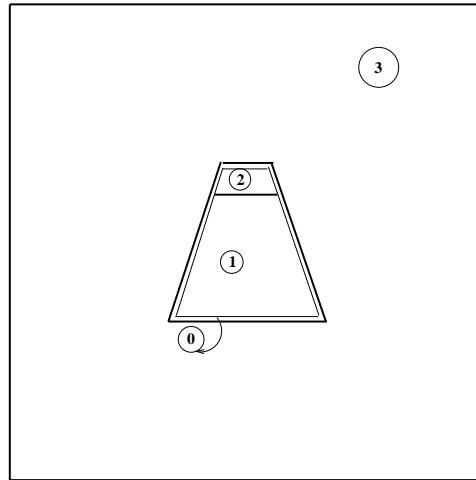
**Fig. 2.** *The experiment*

Two input parameters (data) for the solution of the heat-mass transfer equation in the adsorbent container are obtained as output results from solution of the drying front movement equation:

- quantity of the sublimated water molecules per a time unit, and
- the total time to finalize the process, i.e. the time interval for the parabolic PDE  $[0, T]$ .

The quantity of the sublimated water molecules for a time unit is multiplied by the adsorption heat coefficient in order to obtain the integral intensity of the heater.

We study the heat-mass transfer in the absorption camera via solving the above introduced parabolic boundary value problem. The computational domain (see Fig. 3) is discretised via triangle finite elements (see Fig. 4) using the computer mesh generator **Triangle** ([3, 9]). Let us mention that there are big jumps in the values of the coefficients of the parabolic equation. The computational



**Fig. 3.** The computational domain in the case-study (0 – glass, 1 – zeolite granules, 2 – vacuum, 3 – air)

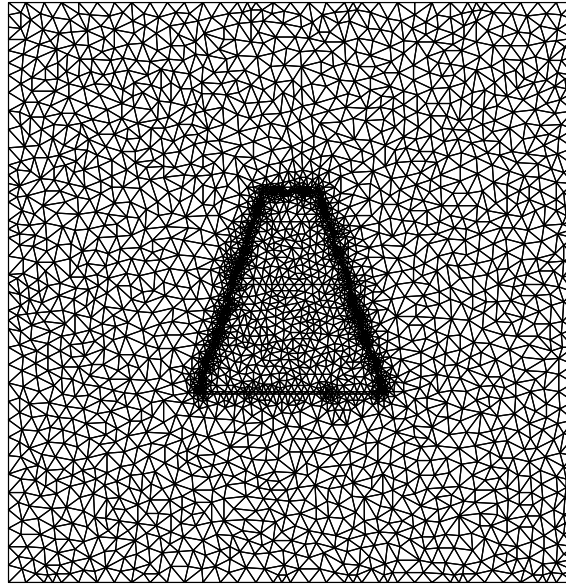
domain consists of four strongly different according to their physical characteristics, subdomains: glass, zeolite granules, vacuum and air. In the beginning of the process the air around the container with zeolite granules is with a room temperature (e.g. about  $20^{\circ}C$ ). A number of computer experiments were done in order to obtain appropriate values of the mesh parameters and the time step. The scalability of the code were studied (see below some results showing the good scalability on several successive grids). Results from one of the provided computer test experiments, where a mesh with 6577 triangles and 3365 nodes and a time step  $\tau = 10sec$ . were used are presented in this paper. The time to end the process of drying  $T$  is an input parameter. CPU time for running the code on Pentium IV, 1.5 GHz is 393.82sec. The temperature field obtained in the adsorption camera can be seen on Fig. 5, a), while on Fig. 5, b) one can see how the zeolite granules are filled in with water molecules coming from the drying container.

## 5 Conclusion

This paper is devoted to a numerical study of a freeze-drying process. Results from some of the performed numerical experiments are presented. In particular, the following subtasks are studied: (i) the drying front movement in the freeze dryer, (ii) the temperature fields in the absorbent camera and (iii) the transfer of the sublimated water molecules and their retention in the sorbent granules. Zeolites granules are used to sublimate the water molecules.

The mathematical model of the process of freeze-drying is described by a system of time-depending differential equations.





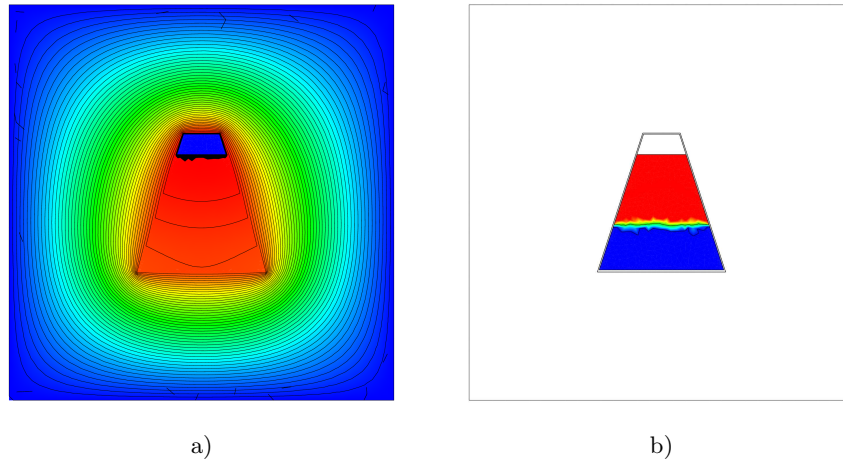
**Fig. 4.** Finite element discretization of the computational domain

For implementation of the presented mathematical algorithm a computer code is developed using the algorithmic languages *Fortran* and *C++*. The scalability of the code was studied on three successive grids and the results are given in Table 1.

**Table 1.** Scalability of the code

Number of unknowns	CPU time	Ratio nodes	Ratio CPU time
$N_1 = 1174$	$T_1 = 137.14$	$N_2/N_1 = 1.85$	$T_2/T_1 = 1.91$
$N_2 = 2171$	$T_2 = 261.52$	$N_3/N_2 = 1.55$	$T_3/T_2 = 1.51$
$N_3 = 3365$	$T_3 = 393.82$	$N_3/N_1 = 2.866$	$T_3/T_1 = 2.872$

The output results of the numerical tests performed with the computer code realizing the presented algorithm are compared with measurements coming from a real-life experiment. These comparisons show that the solver could be successfully used for simulation of the process of freezy-drying.



**Fig. 5.** The temperature field—*a*) and the adsorption of the water molecules—*b*), in the adsorption camera.

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