The parametric analysis of self-freezing in an initially wet porous medium

Y. C. Fey and M. A. Boles
Department of Mechanical and Aerospace Engineering, North Carolina State University,
Raleigh, NC 27695-7910, USA

Received 21 May 1987 and accepted for publication 17 November 1987

The problem of freeze-drying during the self-freezing of a wet porous medium where the freezing and sublimation drying processes occur simultaneously is studied. Analytical solutions of self-freezing in an initially wet semi-infinite porous region are developed by the similarity variable method. The sublimation interface temperature, pressure, and vapor concentration are determined in addition to the positions of the freezing and sublimation interfaces. The vapor transfer in the dried region is modeled by the application of Darcy's and Fick's laws coupled with an additional model for the pressure in this region. For the self-freezing process, the freezing rate is found to be much faster than the sublimation rate. Also, the effects of significant parameters on the sublimation and freezing interface constants are presented for in-depth understanding and application of self-freezing process.

Keywords: heat and mass transfer; porous media; sublimation; filtration

Introduction

The vacuum sublimation process has numerous applications in the food, medical, and chemical industries for freeze-drying heat-sensitive products. In that process, the freezing and sublimation of the substance take place separately, and the porous material is dried in a vacuum after the freezing process is finished. However, in the self-freezing process, the initially wet porous medium is frozen by itself due to vaporization of the bound wet moisture in the vacuum. This results in a temperature drop of the bound substance so that the freezing and sublimation processes occur simultaneously. Another application of the self-freezing process is the vacuum-freezing of products such as food. In the usual sublimation process, the surface temperature is higher than the initial temperature of the medium, which causes heat to flow toward the sublimation interface; but in the self-freezing process the surface temperature is lower than the initial temperature so that the heat flux is transferred to the outer surface. This reversal of the heat transfer direction results in a higher thermal efficiency of the self-freezing process; thus, it is desirable to make this dehydration process or vacuum-freezing process applicable in practice.

An analytical and experimental study of the self-freezing of the porous medium was completed by Hayashi, Komori, and Katayama[1]. For the theoretical model, the above-mentioned investigators only considered the energy portion of the problem; that is, they did not take into account the mass concentration and pressure fields in the dried region. Also, they assumed that the sublimation temperature was at some known constant value which was provided experimentally and the freezing temperature was at 0°C for substance of water. Hence in this work, we consider not only the energy equations of the process but also the equations describing mass concentration and pressure fields in the dried region. Furthermore, we have analytically obtained the temperature, pressure, and vapor concentration at the sublimation interface and determined the positions of the sublimation and freezing interfaces. Since it is of more interest to know the effects of various parameters on both the sublimation and the freezing rates, this paper presents figures to illustrate the variations of the two interface positions with significant parameters, and discusses the physical trends involved in the self-freezing process.

Statement of the problem

An initially wet porous semi-infinite medium is exposed to an environment where the temperature, pressure, and vapor concentration are maintained below the triple point of the bound substance. The porous medium is assumed to be composed of very small solid particles of the same size. Also, the medium is isotropic, homogeneous, rigid, and initially filled with a bound liquid substance. Figure 1 shows a semi-infinite wet porous medium where the temperature and moisture content are initially constant throughout the medium, and the initial temperature is higher than that of the environment. At time greater than zero the temperature, vapor concentration, and pressure of the surface at $x=0$ are maintained at constant

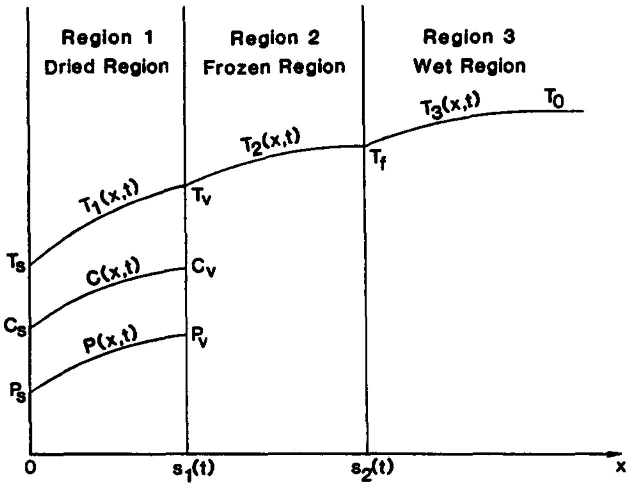


Figure 1 Schematic description for the self-freezing problem

low values, and the self-freezing process begins. When the self-freezing process begins, two transformations (i.e., freezing and sublimation) exist and divide the medium into three regions: dried region, frozen region, and wet region. The moving sublimation and freezing interfaces are assumed sharply thin and located by $x=s_1(t)$ and $x=s_2(t)$, respectively. The temperature, vapor concentration, and pressure at the sublimation interface are in equilibrium and unknown, but are assumed to have constant values which will be determined later. The wet and frozen regions maintain their uniform moisture contents, and no moisture movements occur; therefore, we only consider the heat conduction equations in these two regions. However, in the dried region the vapor transfers outwardly as a result of the interactions of temperature, vapor concentration, and pressure gradients; hence, we consider the energy equation, including the heat convection term, the mass transport equation due to the concentration gradient (Fick's law) and the pressure gradient (Darcy's law), and the equation of the pressure field in the dried region which are used in Refs. 2 and 3.

To formulate the theoretical model of the self-freezing process, we assume that

1. A one-dimensional heat and mass transfer is considered.
2. The radiative heat transfer, thermal expansion of the medium, condensation, Soret and Dufour effects are small and negligible.
3. The thermofluid properties remain constant but may be different for different regions.
4. The Clapeyron equation and ideal-gas law are valid at the sublimation interface.

According to the above bases the self-freezing process may be formulated by the following equations:

$$(\rho C_p)_1 \frac{\partial T_1(x,t)}{\partial t} = K_1 \frac{\partial^2 T_1(x,t)}{\partial x^2} + C_{pw}\dot{W}\frac{\partial T_1(x,t)}{\partial x}, \qquad 0<x<s_1(t) \tag{1}$$

$$(\rho C_p)_2 \frac{\partial T_2(x,t)}{\partial t} = K_2 \frac{\partial^2 T_2(x,t)}{\partial x^2}, \qquad s_1(t)<x<s_2(t) \tag{2}$$

$$(\rho C_p)_3 \frac{\partial T_3(x,t)}{\partial t} = K_3 \frac{\partial^2 T_3(x,t)}{\partial x^2}, \qquad s_2(t)<x<\infty \tag{3}$$

$$\varepsilon \frac{\partial C(x,t)}{\partial t} = \alpha_m \frac{\partial^2 C(x,t)}{\partial x^2} + \kappa \frac{\partial^2 P(x,t)}{\partial x^2}, \qquad 0<x<s_1(t) \tag{4}$$

$$\frac{\partial P(x,t)}{\partial t} = \alpha_p \frac{\partial^2 P(x,t)}{\partial x^2}, \qquad 0<x<s_1(t) \tag{5}$$

Equations 1–3 describe the temperature distributions in the dried, frozen, and wet regions, respectively. The second term on the right side of Equation 1 represents the heat convection due to vapor flow. The vapor flow rate $\dot{W}$ is

$$\dot{W} = \varepsilon M_m C_0 \frac{ds_1(t)}{dt} \tag{6}$$

Equations 4 and 5, which are based upon the Luikov system[4,5] and used in Refs. 2 and 3, describe the concentration and pressure fields, respectively, in the dried region. We also note that the first and second terms on the right side of Equation

Notation

b	Heat convection parameter
C	Molar concentration of moisture
C_0	Molar concentration of frozen bound substance
C_p	Specific heat
C_{pw}	Specific heat of vapor
$\bar{C}$	C/C_{tr}, nondimensional molar concentration of vapor
$\bar{C}_0$	C_0/C_{tr}, nondimensional molar concentration of frozen bound substance
K	Effective thermal conductivity
K_{ij}	K_i/K_j, thermal conductivity ratio
l_1	Latent heat of sublimation
l_2	Latent heat of freezing
L	l_1/R_0T_{tr}, nondimensional parameter
Lu	α_m/α_1, Luikov moisture diffusivity
$\mathrm{Lu_p}$	α_p/α_1, Luikov filtration diffusivity
M_m	Molecular weight of bound substance
P	Pressure
$\bar{p}$	P/P_{tr}, nondimensional pressure
Q_1	$C_{tr}M_m\alpha_1l_1/T_{tr}K_2$, nondimensional latent heat of sublimation
Q_2	$C_{tr}M_m\alpha_1l_2/T_{tr}K_2$, nondimensional latent heat of freezing
R	$C_{tr}R_0T_{tr}/P_{tr}$, nondimensional gas constant
R_0	Universal gas constant
$s_1(t)$	Position of sublimation interface
$s_2(t)$	Position of freezing interface
t	Time
T	Temperature
T_0	Initial temperature
$\dot{W}$	Mass flowrate of vapor
x	Space coordinate
$Z(\eta)$	$P(\eta)+\beta C(\eta)$, transformation variable

Greek symbols

α	Effective thermal diffusivity
α_m	Effective moisture diffusivity
α_p	Filtration motion diffusion coefficient of vapor
α_{ij}	α_i/α_j, thermal diffusivity ratio
β	$(\alpha_m-\varepsilon\alpha_p)/\kappa$
Δ	$\varepsilon\kappa P_{tr}/(\varepsilon\alpha_p-\alpha_m)C_{tr}$ or $-\varepsilon P_{tr}/\beta C_{tr}$, nondimensional permeability
ε	Porosity
η	$x/2\sqrt{\alpha_1 t}$, similarity variable
θ	T/T_{tr}, nondimensional temperature
θ_0	T_0/T_{tr}, nondimensional initial temperature
κ	Permeability
λ_1	$s_1(t)/2\sqrt{\alpha_1 t}$, nondimensional position of sublimation interface
λ_2	$s_2(t)/2\sqrt{\alpha_1 t}$, nondimensional position of freezing interface
ρ	Density

Subscripts

1	Dried region, $0<x<s_1(t)$
2	Frozen region, $s_1(t)<x<s_2(t)$
3	Wet region, $s_2(t)<x<\infty$
f	At freezing interface, $x=s_2(t)$
s	At surface, $x=0$
tr	At triple point of bound substance
v	At sublimation interface, $x=s_1(t)$

4 represent the Fick and Darcy flows, respectively. Equation 5 results from experimental evidence of Luikov[6], which shows that the total pressure gradient is the controlling factor in the transfer of water vapor during the freeze-drying process. The coefficients α_m and α_p used in Equations 4 and 5 are defined by Luikov[4,5]. The boundary and initial conditions are

$$T_3(x, 0) = T_3(\infty, t) = T_0 \tag{7}$$

$$T_1(0, t) = T_s \tag{8}$$

$$C(0, t) = C_s \tag{9}$$

$$P(0, t) = P_s \tag{10}$$

At the sublimation and freezing interfaces the conditions are

$$T_1(s_1, t) = T_2(s_1, t) = T_v \tag{11}$$

$$T_2(s_2, t) = T_3(s_2, t) = T_f \tag{12}$$

$$C(s_1, t) = C_v \tag{13}$$

$$P(s_1, t) = P_v \tag{14}$$

where T_v, C_v, and P_v are the temperature, molar concentration, and pressure, respectively, at the sublimation interface, which are the unknown constants, and T_f is the freezing interface temperature having the known constant value, for substance of water, which is at 0°C.

The energy and moisture balances at the sublimation and freezing interfaces are

$$K_2 \frac{\partial T_2(s_1, t)}{\partial x} - K_1 \frac{\partial T_1(s_1, t)}{\partial x} = \varepsilon C_0 M_m l_1 \frac{ds_1(t)}{dt} \tag{15}$$

$$K_2 \frac{\partial T_2(s_2, t)}{\partial x} - K_3 \frac{\partial T_3(s_2, t)}{\partial x} = \varepsilon C_0 M_m l_2 \frac{ds_2(t)}{dt} \tag{16}$$

$$\alpha_m \frac{\partial C(s_1, t)}{\partial x} + \kappa \frac{\partial P(s_1, t)}{\partial x} = \varepsilon (C_0 - C_v) \frac{ds_1(t)}{dt} \tag{17}$$

The Clapeyron equation relating the sublimation latent heat to the interface parameters, from Equation 8 in Ref. 7, is

$$\frac{C_v T_v}{C_{tr} T_{tr}} = \exp\left[\frac{l_1}{R_0}\left(\frac{1}{T_{tr}} - \frac{1}{T_v}\right)\right] \tag{18}$$

By applying the ideal-gas law at the sublimation interface, we have

$$P_v = C_v R_0 T_v \tag{19}$$

Solution of the problem

Following a similar procedure as in Ref. 3, we introduce the dimensionless similarity variable

$$\eta = x/2\sqrt{\alpha_1 t} \tag{20}$$

into Equations 1–17 and define a new variable $Z(\eta)$ as in Equation 21, which decouples Equations 4 and 5:

$$Z(\eta) = P(\eta) + \beta C(\eta) \tag{21}$$

where

$$\beta = (\alpha_m - \varepsilon \alpha_p)/\kappa \tag{22}$$

The locations of the sublimation and freezing interfaces are assumed to be given, respectively, by

$$s_1(t) = 2\lambda_1 \sqrt{\alpha_1 t} \tag{23}$$

and

$$s_2(t) = 2\lambda_2 \sqrt{\alpha_1 t} \tag{24}$$

where the nondimensional sublimation and freezing interface positions λ_1, λ_2 are the unknown constants to be determined during the solution.

With the introduction of the new variables η, λ_1, and λ_2, we note that the dried region corresponds to $0 < \eta < \lambda_1$, the frozen region corresponds to $\lambda_1 < \eta < \lambda_2$, and the wet region corresponds to $\lambda_2 < \eta < \infty$. The problem is transformed to the following system of ordinary differential equations with variable coefficients subject to the transformed boundary and interface conditions:

$$\frac{d^2 T_1(\eta)}{d\eta^2} + 2(b\lambda_1 + \eta) \frac{dT_1(\eta)}{d\eta} = 0, \qquad 0 < \eta < \lambda_1 \tag{25}$$

$$\frac{d^2 T_2(\eta)}{d\eta^2} + 2\frac{\alpha_1}{\alpha_2} \eta \frac{dT_2(\eta)}{d\eta} = 0, \qquad \lambda_1 < \eta < \lambda_2 \tag{26}$$

$$\frac{d^2 T_3(\eta)}{d\eta^2} + 2\frac{\alpha_1}{\alpha_3} \eta \frac{dT_3(\eta)}{d\eta} = 0, \qquad \lambda_2 < \eta < \infty \tag{27}$$

$$\frac{d^2 P(\eta)}{d\eta^2} + 2\frac{\alpha_1}{\alpha_p} \eta \frac{dP(\eta)}{d\eta} = 0, \qquad 0 < \eta < \lambda_1 \tag{28}$$

$$\frac{d^2 Z(\eta)}{d\eta^2} + 2\varepsilon\frac{\alpha_1}{\alpha_m} \eta \frac{dZ(\eta)}{d\eta} = 0, \qquad 0 < \eta < \lambda_1 \tag{29}$$

where the heat convection parameter b in Equation 25 is

$$b = \varepsilon M_m C_{pw} C_0 \alpha_1 / K_1 \tag{30}$$

which shows the existence and effect of the heat convection. The system of Equations 25–29 can be solved exactly for T_1, T_2, T_3, P, and Z with the transformed boundary and interface conditions, and the solution for $C(\eta)$ is obtained from Equation 21. After substituting the above solutions into the interface equations and performing the required manipulations, we get the five transcendental interface equations. By using the nondimensional parameters defined in the notation, we write the solutions as follows:

$$\theta_1(\eta) = \theta_s + (\theta_v - \theta_s) \frac{\operatorname{erfc}(b\lambda_1) - \operatorname{erfc}(\eta + b\lambda_1)}{\operatorname{erfc}(b\lambda_1) - \operatorname{erfc}(\lambda_1 + b\lambda_1)}, \qquad 0 < \eta < \lambda_1 \tag{31}$$

$$\theta_2(\eta) = \theta_v + (\theta_f - \theta_v) \frac{\operatorname{erfc}(\sqrt{\alpha_{12}}\,\lambda_1) - \operatorname{erfc}(\sqrt{\alpha_{12}}\,\eta)}{\operatorname{erfc}(\sqrt{\alpha_{12}}\,\lambda_1) - \operatorname{erfc}(\sqrt{\alpha_{12}}\,\lambda_2)}, \qquad \lambda_1 < \eta < \lambda_2 \tag{32}$$

$$\theta_3(\eta) = \theta_0 + (\theta_f - \theta_0) \frac{\operatorname{erfc}(\sqrt{\alpha_{13}}\,\eta)}{\operatorname{erfc}(\sqrt{\alpha_{13}}\,\lambda_2)}, \qquad \lambda_2 < \eta < \infty \tag{33}$$

$$\bar{p}(\eta) = \bar{p}_s + (\bar{p}_v - \bar{p}_s) \frac{\operatorname{erf}(\eta/\sqrt{\mathrm{Lu_p}})}{\operatorname{erf}(\lambda_1/\sqrt{\mathrm{Lu_p}})}, \qquad 0 < \eta < \lambda_1 \tag{34}$$

$$\begin{aligned} \bar{C}(\eta) = \bar{C}_s &+ \left[\frac{\Delta}{\varepsilon}(\bar{p}_s - \bar{p}_v) + (\bar{C}_v - \bar{C}_s)\right] \frac{\operatorname{erf}(\sqrt{\varepsilon/\mathrm{Lu}}\,\eta)}{\operatorname{erf}(\sqrt{\varepsilon/\mathrm{Lu}}\,\lambda_1)} \\ &+ \frac{\Delta}{\varepsilon}(\bar{p}_v - \bar{p}_s) \frac{\operatorname{erf}(\eta/\sqrt{\mathrm{Lu_p}})}{\operatorname{erf}(\lambda_1/\sqrt{\mathrm{Lu_p}})}, \qquad 0 < \eta < \lambda_1 \end{aligned} \tag{35}$$

The energy and moisture balances at the sublimation interface become

$$\frac{\theta_f - \theta_v}{\theta_v - \theta_s} \frac{\sqrt{\alpha_{12}} \exp(-\alpha_{12}\lambda_1^2)}{\operatorname{erfc}(\sqrt{\alpha_{12}}\,\lambda_1) - \operatorname{erfc}(\sqrt{\alpha_{12}}\,\lambda_2)} - \frac{K_{12} \exp\{-(\lambda_1 + b\lambda_1)^2\}}{\operatorname{erfc}(b\lambda_1) - \operatorname{erfc}(\lambda_1 + b\lambda_1)} = \frac{\sqrt{\pi}\,\varepsilon \bar{C}_0 Q_1 \lambda_1}{\theta_v - \theta_s} \tag{36}$$

and

$$\frac{\sqrt{\mathrm{Lu}/\varepsilon}[\Delta(\bar{p}_s-\bar{p}_v)+\varepsilon(\bar{C}_v-\bar{C}_s)]\exp(-\varepsilon\lambda_1^2/\mathrm{Lu})}{\mathrm{erf}(\lambda_1/\sqrt{\mathrm{Lu}/\varepsilon})} + \frac{\Delta(\bar{p}_v-\bar{p}_s)\sqrt{\mathrm{Lu}_p}\exp(\lambda_1^2/\mathrm{Lu}_p)}{\mathrm{erf}(\lambda_1/\sqrt{\mathrm{Lu}_p})} = \sqrt{\pi}\,\lambda_1\varepsilon(\bar{C}_0-\bar{C}_v) \tag{37}$$

The energy balance at the freezing interface becomes

$$\frac{\theta_f-\theta_v}{\theta_0-\theta_f}\frac{\sqrt{\alpha_{12}}\exp(-\alpha_{12}\lambda_2^2)}{\mathrm{erfc}(\sqrt{\alpha_{12}}\,\lambda_1)-\mathrm{erfc}(\sqrt{\alpha_{12}}\,\lambda_2)} - \frac{K_{32}\sqrt{\alpha_{13}}\exp(-\alpha_{13}\lambda_2^2)}{\mathrm{erfc}(\sqrt{\alpha_{13}}\,\lambda_2)} = \frac{\sqrt{\pi}\,\varepsilon\bar{C}_0Q_2\lambda_2}{\theta_0-\theta_f} \tag{38}$$

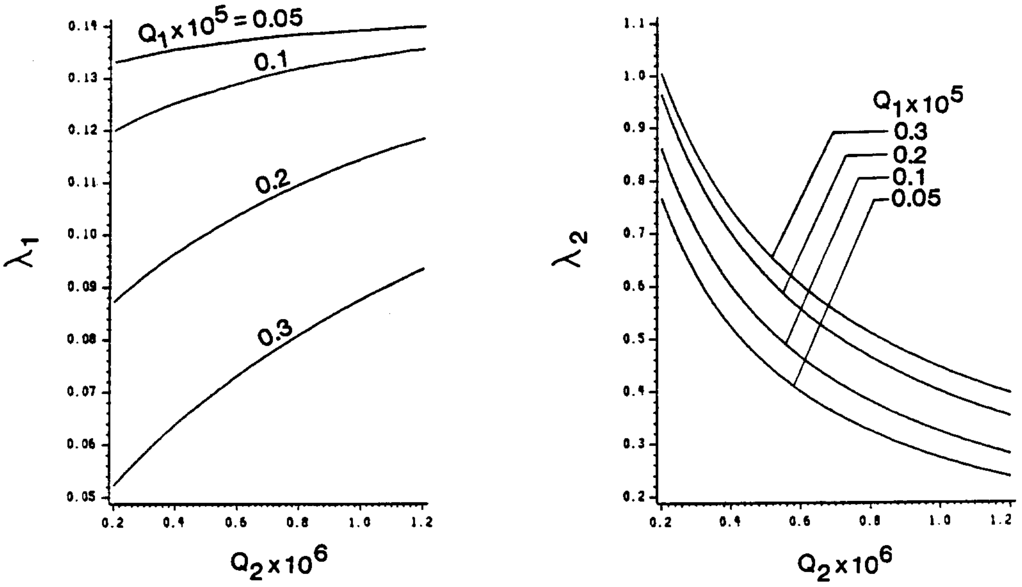
The Clapeyron equation becomes

$$\bar{C}_v\theta_v = \exp[L(1-1/\theta_v)] \tag{39}$$

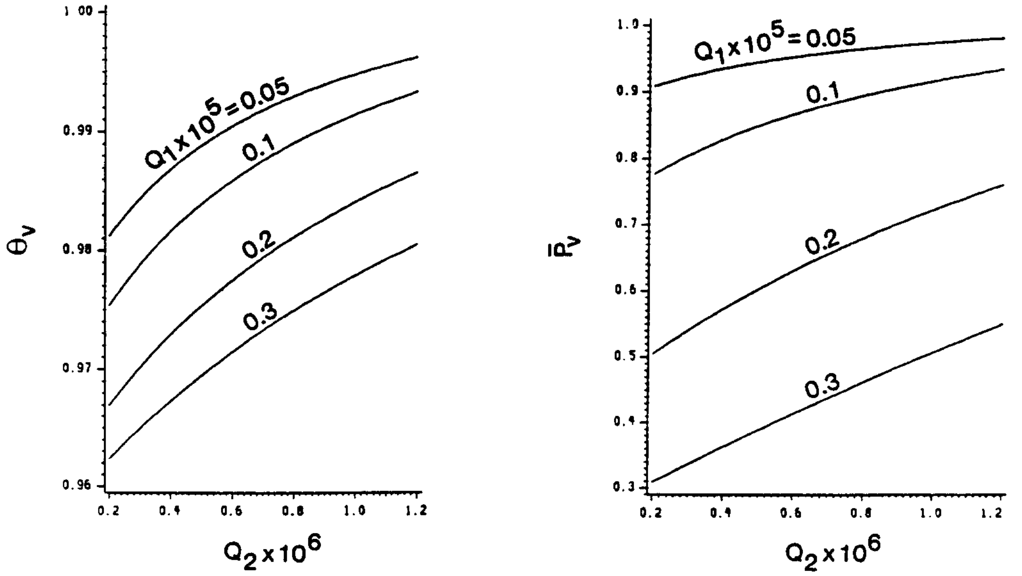
and the ideal-gas law becomes

$$\bar{p}_v = \bar{C}_v R\theta_v \tag{40}$$

The nondimensional sublimation interface position λ_1, freezing interface position λ_2, nondimensional sublimation interface temperature θ_v, pressure $\bar{p}_v$, and molar concentration $\bar{C}_v$ are then obtained by numerically solving the simultaneous equations 36–40. Once the interface constants are known, Equations 31–35 readily yield the exact solution to the self-freezing problem.

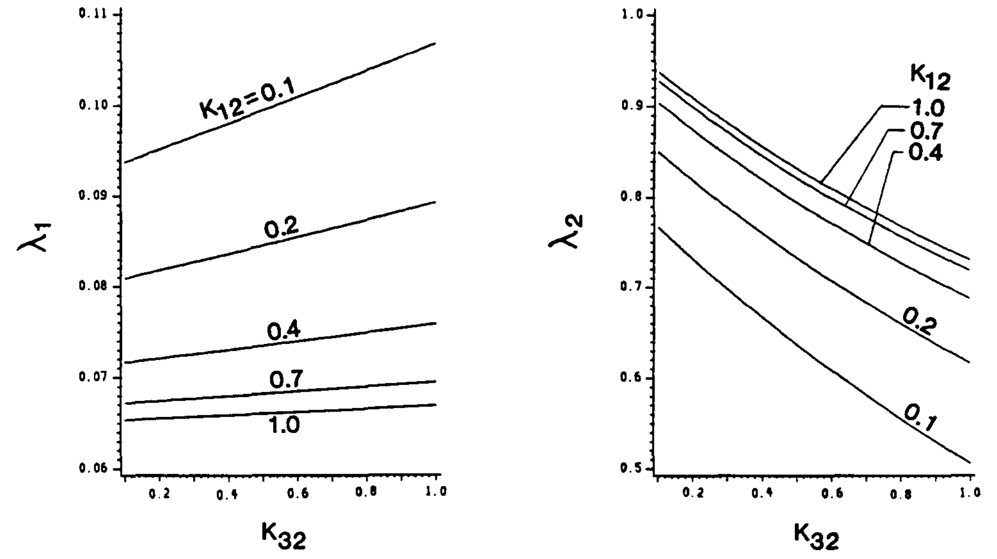


(a) Sublimation interface position (b) Freezing interface position



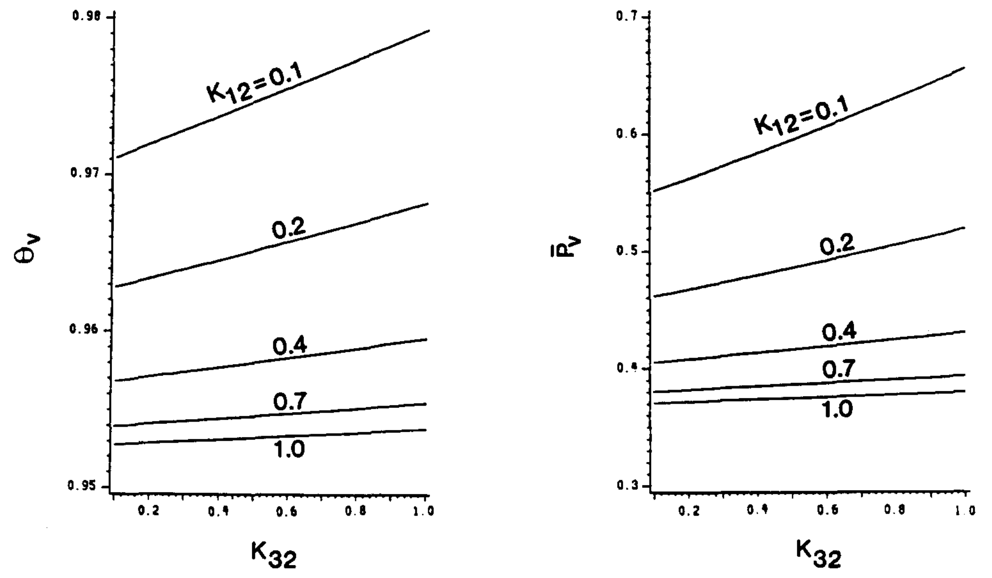
(c) Sublimation interface temperature (d) Sublimation interface pressure

Figure 2 Effects of nondimensional latent heat of sublimation and freezing on nondimensional sublimation interface position, freezing interface position, and sublimation interface temperature and pressure



(a) Sublimation interface position

(b) Freezing interface position



(c) Sublimation interface temperature

(d) Sublimation interface pressure

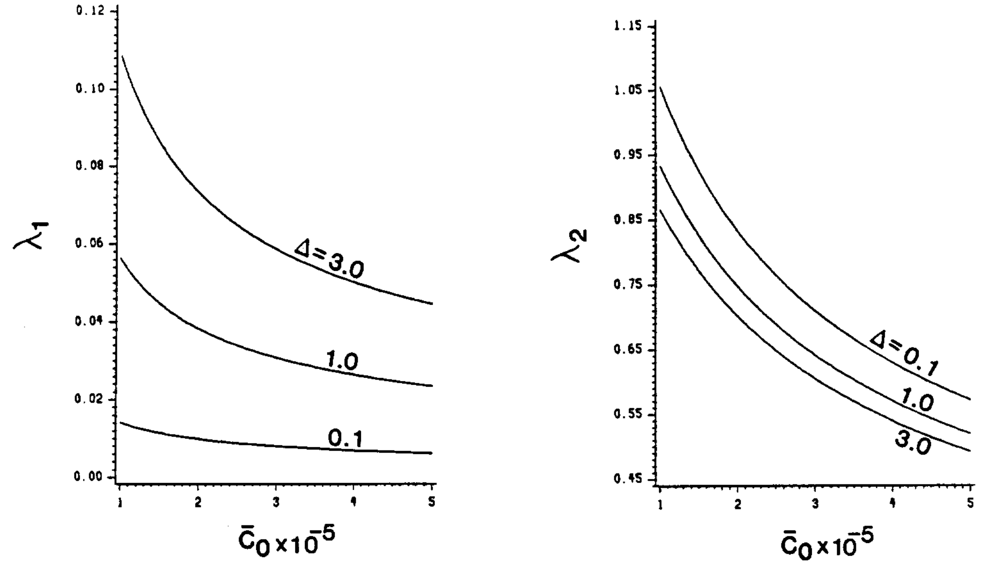
Figure 3 Effects of thermal conductivity ratios K_{12} and K_{32} on nondimensional sublimation interface position, freezing interface position, and sublimation interface temperature and pressure

Results and discussion

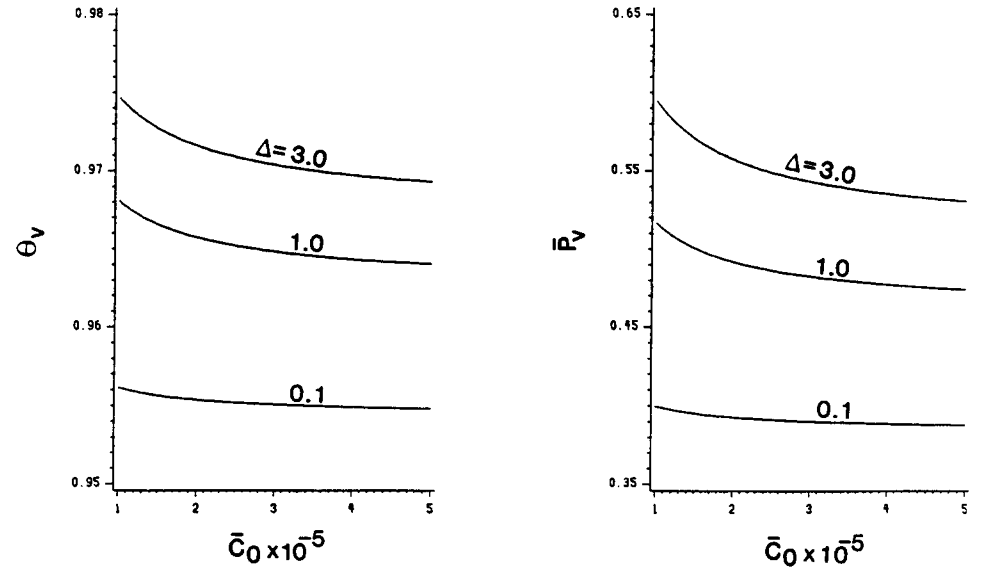
The self-freezing in a porous medium involving three phases and two transformations (i.e., freezing and sublimation) is a complicated physical process in which both the heat flux and mass flux transfer outwardly from the sublimation interface. Several parameters are found to have different effects on the self-freezing process from those on the usual vacuum sublimation process. These effects result in the reversal of the movement of the positions of sublimation and freezing interfaces in the self-freezing process.

To better understand the self-freezing process, sample calculations are performed to illustrate the physical trends of the effects of various system parameters on the sublimation and freezing interface parameters for the water-glass powder medium used in Ref. 1. The sublimation interface speed is proportional to the nondimensional constant λ_1, and the freezing interface speed is proportional to the nondimensional constant λ_2. On the figures presented in this study, only the parameters having values different from the reference values are indicated. The reference thermal properties used in this study are taken from Ref. 1, and the reference filtration properties are obtained from Ref. 4. The selected reference values are $b=0.7$, $\Delta=5.0$, $K_{12}=0.1$, $K_{32}=0.3$, $\alpha_{12}=0.2$, $\alpha_{13}=0.6$, $\varepsilon=0.3$, $\mathrm{Lu}=0.1$, $\bar{C}_0=2.0\times10^5$, $\bar{C}_s=0.2$, $\theta_0=1.02$, $\theta_s=0.95$, $\bar{p}_s=0.2$, $R=1.0$, $\mathrm{Lu}_p=600$, $L=20$, $Q_1=0.2\times10^{-5}$, $Q_2=0.4\times10^{-6}$, and $\theta_f=1.0$.

Figure 2 illustrates the effects of the nondimensional latent heat of sublimation and freezing on the nondimensional sublimation interface position λ_1, freezing interface position λ_2, and sublimation interface temperature and pressure. As the sublimation latent heat Q_1 increases, more energy is required to sublimate the material. Also, the interface temperature and interface pressure decrease with increasing latent heat Q_1. This



(a) Sublimation interface position **(b) Freezing interface position**

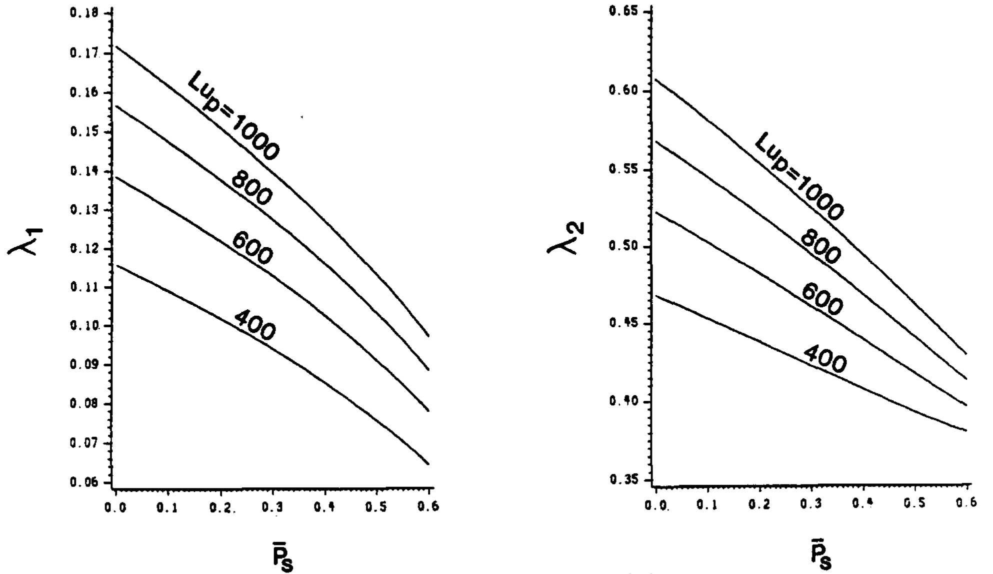


(c) Sublimation interface temperature **(d) Sublimation interface pressure**

Figure 4 Effects of nondimensional permeability and molar concentration of frozen bound substance on nondimensional sublimation interface position, freezing interface position, and sublimation interface temperature and pressure

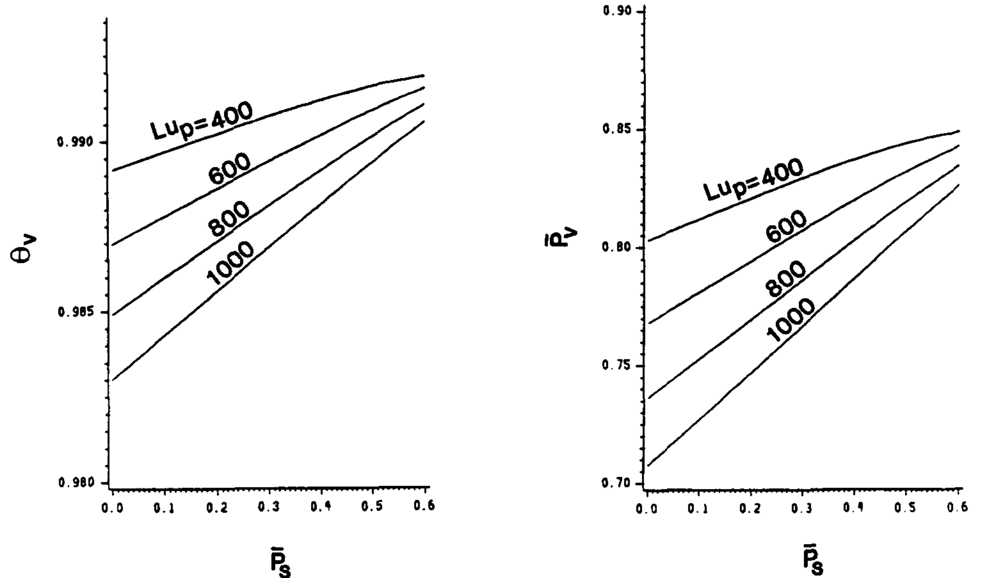
results in a lower pressure gradient across the dried region and in a higher temperature gradient across the frozen region. Accordingly, increasing the sublimation latent heat Q_1 decreases the sublimation speed and increases freezing speed. On the other hand, the sublimation speed increases and the freezing speed decreases when the latent heat of freezing Q_2 increases. The substance having the higher latent heat of freezing will release more energy per unit mass, which results in lower freezing interface speed, higher temperature in the frozen layer, and higher sublimation interface temperature and pressure. The energy released by the freezing material is conducted through the frozen material and made available to the sublimation process, which increases the sublimation interface speed. The sublimation latent heat has more significant effect on λ_1 than on λ_2; however, the freezing latent heat has significant effect on both λ_1 and λ_2.

Figure 3 illustrates the effects of the nondimensional thermal conductivity ratios K_{12} and K_{32} on the nondimensional sublimation interface position λ_1, freezing interface position λ_2, and sublimation interface temperature and pressure. Decreasing K_{12} causes less heat flux to be conducted from the sublimation interface through the dried region, which results in a higher sublimation interface temperature and pressure. Thus, more energy is available for sublimation, and the higher pressure gradient also aids the sublimation process. The higher sublimation temperature results in a small heat flux conducted from the freezing interface, which results in a lower freezing rate. The figure also shows that a higher sublimation interface speed and lower freezing interface speed occur for the medium with larger thermal conductivity K_3 (i.e., higher K_{32}). Since more energy is conducted toward the freezing interface through the wet layer due to large K_3, the lower freezing interface speed results. However, with larger values of K_{32}, the sublimation interface has higher temperature and pressure values, which



(a) Sublimation interface position

(b) Freezing interface position



(c) Sublimation interface temperature

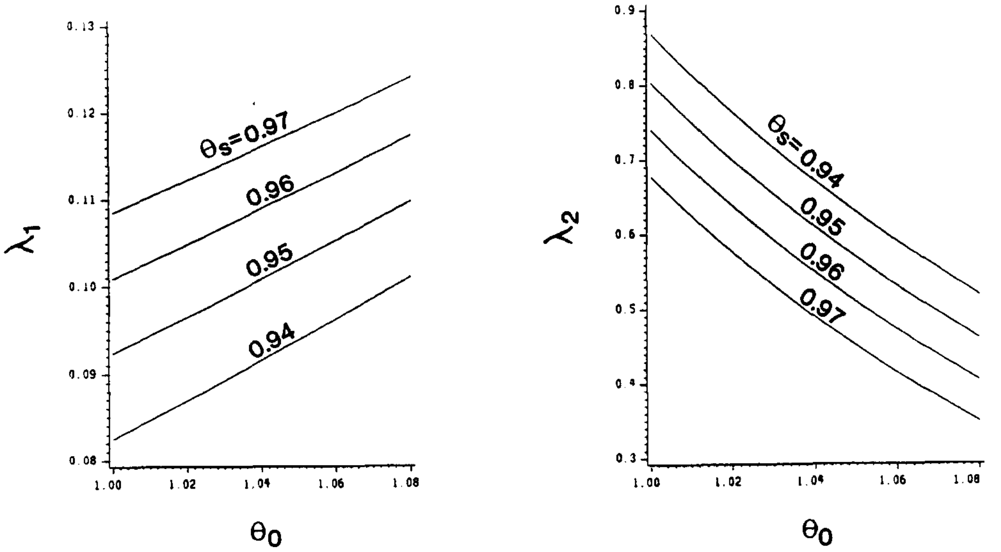
(d) Sublimation interface pressure

Figure 5 Effects of Luikov filtration diffusivity and nondimensional surface pressure on nondimensional sublimation interface position, freezing interface position, and sublimation interface temperature and pressure for θ_s=0.985

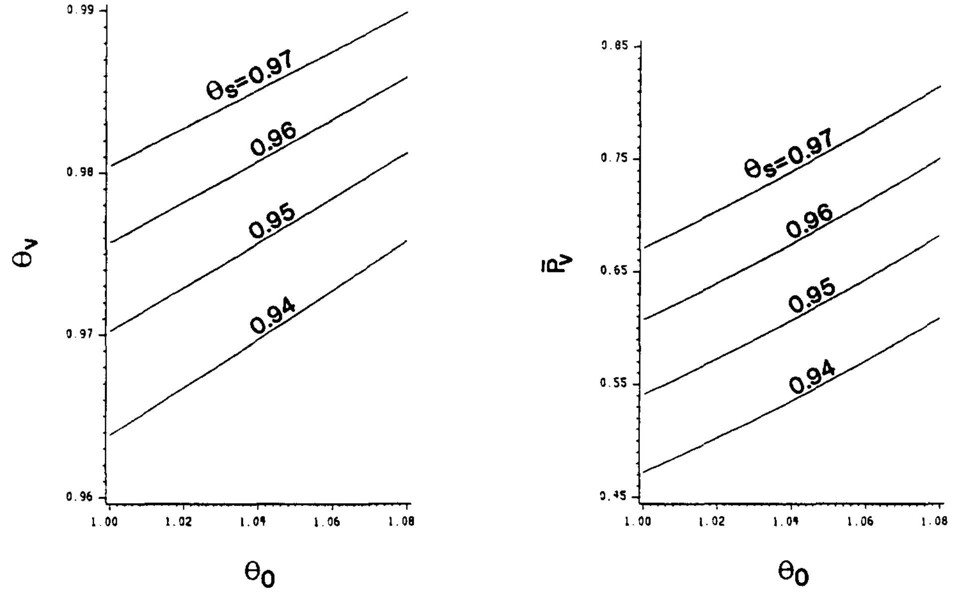
explains the higher sublimation speed. We note that the rates of change in λ_1 and λ_2 with K_{12} are significant as K_{12} decreases.

Figure 4 shows the effects of the nondimensional permeability and molar concentration of frozen bound substance on the nondimensional sublimation interface position λ_1, freezing interface position λ_2, and sublimation interface temperature and pressure. The porous medium with a higher permeability has a lower resistance to vapor flow through the dried region and has higher values of sublimation interface temperature and pressure; thus, the medium is more easily sublimated, and a thicker dried region exists. The resistance for conduction heat transfer through the dried region is increased, and most of the energy released by the freezing interface is used for sublimation. The result is a higher sublimation interface speed and a lower freezing interface speed as the permeability increases. The figure also shows that the sublimation and freezing interface speeds decrease as the molar concentration of frozen bound substance $\bar{C}_0$ increases. This result can be explained by the fact that, as $\bar{C}_0$ increases, more moles of substance need be sublimated at the sublimation interface, which requires more energy for sublimation and reduces the sublimation speed. However, at the freezing interface more latent heat of freezing must be released per unit volume, and the freezing interface speed is slowed. We point out that the effect of $\bar{C}_0$ on λ_1 is more important for higher values of permeability.

Figure 5 illustrates the effects of the Luikov filtration diffusivity, $\mathrm{Lu_p}$, and nondimensional surface pressure, $\bar{P}_s$, on the nondimensional sublimation interface position λ_1, freezing interface position λ_2, and sublimation interface temperature and pressure for $\theta_s=0.985$. As $\mathrm{Lu_p}$ increases, both the sublimation and freezing interface speeds increase. The quantity $\mathrm{Lu_p}$ is a measure of the speed of propagation of pressure waves through the porous medium; thus, higher sublimation rates will be expected for larger values of $\mathrm{Lu_p}$ regardless of the lower interface



(a) Sublimation interface position **(b) Freezing interface position**



(c) Sublimation interface temperature **(d) Sublimation interface pressure**

Figure 6 Effects of nondimensional surface temperature and initial temperature on nondimensional sublimation interface position, freezing interface position, and sublimation interface temperature and pressure

pressure. With increased Lu_p larger amounts of energy are consumed at the sublimation interface, and a lower sublimation interface temperature and a higher heat flux through the frozen region result. This explains the faster freezing interface speed with larger Lu_p. Moreover, when the surface pressure increases, lower pressure gradients exist in the dried region, which give rise to a slower sublimation rate. This in turn slows the freezing interface speed. Note that the surface pressure has a significant influence on the sublimation and freezing interface speeds.

Figure 6 shows the effects of the nondimensional surface temperature, θ_s, and initial temperature, θ_0, on the nondimensional sublimation interface position λ_1, freezing interface position λ_2, and sublimation interface temperature and pressure. For the higher surface temperature, the heat flux conducted from the sublimation interface is reduced, and more energy is available for sublimation. Also due to the higher sublimation interface temperature and pressure, the heat flux is smaller in the higher temperature frozen layer, and the substance is easier to sublimate. Thus, increasing the surface temperature results in a higher sublimation interface speed and lower freezing interface speed. However, when the initial temperature increases, the heat flux toward the freezing interface is increased due to the larger temperature difference in the wet region, which slows down the freezing interface speed. The heat flux from the freezing interface is conducted through the frozen layer, which is now at a higher temperature and interface pressure. Thus, increasing the initial temperature increases the sublimation interface speed and decreases the freezing interface speed. Note that the surface and initial temperatures have significant effects on λ_1 and λ_2.

Results for other parameters, such as thermal diffusivity ratios α_{12} and α_{13}, porosity ε, Luikov number Lu, and heat convection parameter b, have also been obtained. We now briefly describe their effects without the accompanying figures as follows: Increasing the thermal diffusivity ratios α_{12} and α_{13} increases the sublimation interface speed and decreases the freezing interface speed; however, increasing the porosity significantly decreases the sublimation and freezing rates. Also, Lu has a negligible effect on the sublimation and freezing interface speeds. Moreover, although the heat convection parameter b has negligible influence on the sublimation and freezing interface speeds, the results show that the sublimation interface speed increases and the freezing interface speed decreases as b increases.

Conclusion

The following conclusions may be drawn:

(1) The freezing interface speed is much faster than the sublimation interface speed, because the latent heat of sublimation is several times larger than that of freezing.

(2) The self-freezing process basically has two applications in practice; that is, it can be practiced in vacuum-sublimation-drying of heat-sensitive material or in vacuum-freezing of moist material. The effects of physical parameters on the self-freezing process can be sorted into the following groups: (i) The sublimation interface speed λ_1 increases for smaller values in Q_1, K_{12}, $\bar{C}_0$, $\bar{P}_s$, ε, and for higher values in Q_2, K_{32}, Δ, Lu_p, θ_s, θ_0, α_{12}, α_{13}, b. (ii) The freezing interface speed λ_2 increases for higher values in Q_1, K_{12}, Lu_p, and for smaller values in Q_2, K_{32}, Δ, $\bar{C}_0$, $\bar{P}_s$, θ_s, θ_0, α_{12}, α_{13}, ε, b. Furthermore, in the application of vacuum-sublimation-drying, a fast drying rate is desired; therefore, the controllable parameters θ_s, θ_0 should be maintained at higher values, but the surface pressure $\bar{p}_s$ at a lower value. In vacuum-freezing, a fast freezing speed is desired; accordingly all the controllable parameters θ_s, θ_0, $\bar{P}_s$ should be kept at low values.

References

1 Hayashi, Y., Komori, T., and Katayama, K. Analytical and experimental investigation of self-freezing. *J. Heat Transfer* 1975, 321–325

2 Fey, Y. C. and Boles, M. A. An analytical study of the effect of Darcy's and Fick's laws on the sublimation of a frozen semi-infinite porous medium. *J. Heat Transfer* 1987, **109**, 1045–1048

3 Fey, Y. C. and Boles, M. A. An analytical study of the effect of convection heat transfer on the sublimation of a frozen semi-infinite porous medium. *Int. J. Heat Mass Transfer* 1987, **30**, 771–779

4 Luikov, A. V. *Heat and Mass Transfer in Capillary-Porous Bodies*. Pergamon Press, Oxford, 1966

5 Luikov, A. V. Systems of differential equations of heat and mass transfer in capillary-porous bodies (review). *Int. J. Heat Mass Transfer* 1975, **18**, 1–14

6 Luikov, A. V. Heat and mass transfer in freeze-drying at high vacuum. International Institute of Refrigeration, Commission X, Lausanne, Switzerland, 1969

7 Lin, S. An exact solution of the sublimation problem in a porous medium. Part II, with an unknown temperature and vapor concentration at the moving sublimation front. *J. Heat Transfer* 1982, **104**, 808–811