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MATHEMATICAL MODELLING OF MOISTURE DESORPTION IN A POROUS MEDIUM

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SUMMARY

This paper discusses heat and mass transfer in desorption drying. A basic equation system is derived to describe coupled heat and mass transfer in a porous medium with moisture desorption under temperature gradients and a vacuum environment. The desorption mushy zone model is used to obtain an exact solution for coupled heat and mass transfer with a moving desorption mushy zone in a porous half-space. The results are analysed numerically to demonstrate the effects of various parameters on desorption.

KEY WORDS: desorption; porous medium; moving interface

1. INTRODUCTION

In recent years there has been much interest in the physics of porous media in freeze-drying, owing to its extensive application in the food, chemical and medical industries.

A traditional freeze-drying process consists of two successive steps, i.e. sublimation drying and desorption drying. In an ordinary freeze-drying process, desorption drying is designed to begin after sublimation drying comes to an end, by raising the heating temperature and lowering the vacuum pressure.

Most authors have derived the theory of desorption based on Fick's law. The most extensively used desorption drying theory is that of King (1968), which has also been detailed by Mellor (1978). The basic assumptions in King's theory are as follows.

(i) The vapour transfer is described by Fick's law.
(ii) Vapour is in equilibrium with the sorbed moisture.
(iii) The heat of sorption is constant.
(iv) The heat of sorption is greater than the sensible heat.
(v) Surface diffusion of moisture is ignored.
(vi) The amount of sorbed moisture in the liquid phase is much larger than that of moisture in the pores in the vapour phase.

By ignoring the transient terms in the mass and energy conservation equations, he obtained the following equation to describe the desorption drying:

$$\frac{\partial f}{\partial x} = \frac{\partial}{\partial x}\left(D_{\text{eff}}\frac{\partial f}{\partial x}\right) \tag{1}$$

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where D_{eff} is the effective diffusivity and f is the moisture content sorbed on the walls of the porous skeleton.

Obviously, there exist some shortcomings in an equation for moisture movement in desorption drying constructed in this way: firstly, the equation describes the vapour transfer in an indirect way. Vapour transfer is undoubtedly by diffusion, which should only be dependent on vapour concentration gradients in the vapour space — the pores in the porous medium — not on the sorbed moisture content gradients. The desorption of sorbed moisture just provides a mass source for vapour transfer; secondly, the equation implicitly ignores the effect of transient heat and mass transfer terms, although it looks like an unsteady transfer equation; thirdly, and most importantly, the given equation does not apply to the last phase of desorption drying, where the assumption that the amount of sorbed moisture in the condensed phase is much greater than that in the vapour phase breaks down.

In what follows we endeavour to develop a novel desorption drying theory to overcome the above difficulties by extending our recent researcher on heat and mass transfer in simultaneous sublimation and desorption drying (Peng *et al.*, 1992a,b; Peng and Chen, 1993), especially the concept of desorption mushy zone (Peng and Chen, 1994), to ordinary desorption drying. We first develop the coupled heat and mass transfer equations applicable to desorption drying, then derive the exact solution of heat and mass transfer with a desorption mushy zone in a porous half-space to analyse the effect of various parameters on desorption.

2. BASIC EQUATIONS OF DESORPTION DRYING

Desorption drying is essentially a process of coupled heat and mass transfer. Heat transfer through conduction, convection and radiation to the humid porous region — provides energy to desorb the moisture. Heat and mass transfer are coupled through the phase-change — the desorption process. In this process, the temperature, vapour concentration and sorbed moisture content change with space and time.

2.1 Energy conservation

Applying energy conservation to the elements in a porous medium with moisture desorption yields

$$\frac{\partial}{\partial t}(\rho f h_{\mathrm{w}} + C h_{\mathrm{v}} + C_{\mathrm{a}} h_{\mathrm{a}} + C_{\mathrm{s}} h_{\mathrm{s}}) = -\operatorname{div}(q + h_{\mathrm{v}} j_{\mathrm{v}} + h_{\mathrm{a}} j_{\mathrm{a}}) \tag{2}$$

here ρ is the density, f the volumetric fraction of the sorbed moisture, h the enthalpy, C the concentration, q the heat flux, and j the mass flux. The subscripts w, v, a, and s represent the sorbed water, vapour, air and solid skeleton, respectively, but the subscript for vapour concentration is omitted.

In deriving the Luikov equations applicable to sublimation drying, Peng (1994) ignored the desorption of sorbed moisture so that the volumetric fraction of sorbed moisture f was considered constant. Here, we have to consider the change of f with space and time.

Defining the isobaric specific heat by

$$c = \left(\frac{\mathrm{d}h}{\mathrm{d}T}\right)_p \tag{3}$$

then

$$(\rho c)\frac{\partial T}{\partial t} + \rho h_{\mathrm{w}}\frac{\partial f}{\partial t} + h_{\mathrm{v}}\frac{\partial C}{\partial t} + h_{\mathrm{a}}\frac{\partial C_{\mathrm{a}}}{\partial t} = -\operatorname{div} q - (j_{\mathrm{v}} c_{\mathrm{v}} + j_{\mathrm{a}} c_{\mathrm{a}})\nabla T - h_{\mathrm{v}} \operatorname{div} j_{\mathrm{v}} - h_{\mathrm{a}} \operatorname{div} j_{\mathrm{a}} \tag{4}$$

where (ρc) is the effective product of concentration and isobaric heat of the mushy zone

$$(\rho c) = \rho f c_{\mathrm{w}} + C c_{\mathrm{v}} + C_{\mathrm{s}} c_{\mathrm{s}} + C_{\mathrm{a}} c_{\mathrm{a}} \tag{5}$$

2.2 Mass conservation

Then mass conservation of vapour yields

$$\frac{\partial C}{\partial t} = -\operatorname{div} j_{\mathrm{v}} - \rho \frac{\partial f}{\partial t} \tag{6}$$

The mass conservation of inert gas (air) yields

$$\frac{\partial C_{\mathrm{a}}}{\partial t} = -\operatorname{div} j_{\mathrm{a}} \tag{7}$$

Substitution of equations (6) and (7) into equation (4) yields

$$(\rho c)\frac{\partial T}{\partial t} = -\operatorname{div} q - (j_{\mathrm{v}}c_{\mathrm{v}} + j_{\mathrm{a}}c_{\mathrm{a}})\cdot\nabla T + \rho(h_{\mathrm{v}} - h_{\mathrm{w}})\frac{\partial f}{\partial t} \tag{8}$$

The first term on the right-hand side of equation (8) represents the conduction heat transfer, the second term represents the convection heat transfer, and the third term represents the heat sink due to desorption. In freeze-drying, the contribution of convection is very small compared to the conduction heat transfer. (Luikov, 1966, 1975; Peng, 1994), so that the second term can be ignored.

2.3. Constitutive equations

The heat conduction is represented by the Fourier law:

$$q = -k\nabla T \tag{9}$$

where k is the effective heat conductivity for the porous medium.

The mass transfer is represented by Fick's law:

$$j_{\mathrm{v}} = -\alpha_{\mathrm{m}}\nabla C \tag{10a}$$

$$j_{\alpha} = -\alpha_{\mathrm{ma}}\nabla C_{\mathrm{a}} \tag{10b}$$

The so-defined mass diffusional law may be found in the work of Bird *et al.* (1960, pp. 542–546), and is used very often in chemical engineering.

2.4 Heat and mass transfer equations applicable to desorption drying

Writing $h_{\mathrm{v}} - h_{\mathrm{w}} = H_{\mathrm{p}}$, and substituting equations (9) and (10) into equations (6)–(8), we obtain

$$(\rho c)\frac{\partial T}{\partial t} = \operatorname{div}(k\nabla T) + \rho H_{\mathrm{p}}\frac{\partial f}{\partial t} \tag{11a}$$

$$\frac{\partial C}{\partial t} = \operatorname{div}(\alpha_{\mathrm{m}}\nabla C) - \rho\frac{\partial f}{\partial t} \tag{11b}$$

$$\frac{\partial C_{\mathrm{a}}}{\partial t} = \operatorname{div}(\alpha_{\mathrm{ma}}\nabla C_{\mathrm{a}}) \tag{11c}$$

The above equations, together with the corresponding boundary and initial conditions, constitute the complete mathematical description of a desorption drying problem. In contrast to King's theory, the above equations provide a deep insight into the transport mechanism in desorption drying, and the coupling relationship between heat and mass transfer.

Generally speaking, the thermophysical properties in the above equations are functions of temperature, pressure, vapour and air concentrations, so that the equations can only be solved numerically. However, in desorption drying, the temperature and concentration change very slowly, so that the constant thermophysical property analysis as very often carried out in the literature, (e.g. by Luikov, 1975), is still helpful for our understanding of the practical process. In the following section, we present an exact solution of the desorption of a porous half-space to analyse the effect of various parameters.

3. ANALYSIS OF DESORPTION IN A POROUS HALF-SPACE

3.1 Physical and mathematical model

The physical model is shown in Figure. 1. A semi-infinite sublimed porous medium initially has a uniform sorbed moisture content f_0 and uniform temperature T_0. At times greater than zero, it is exposed to a vacuum environment, and its surface is kept at constant temperature T_s, which is higher than the desorption temperature T_d. As a result, the desorption process begins, and the whole region is divided into two regions, namely, the desorbing region and the dried region. Furthermore, as indicated in our recent work (Peng and Chen, 1994), on freeze-drying, the pressure gradient in the porous sublimed region is very small, so that we can consider desorption as approximately an isobaric process where the desorption occurs over an extended temperature range (the desorption mushy zone)

Based on the preceding theory, and the following assumptions:

(i) the problem is one dimensional;
(ii) when the temperature is higher than the desorption temperature T_d, the sorbed moisture content reduces to f_d, which is the moisture content resulting from the combined effects of chemisorption, static electric forces, etc., which cannot be desorbed in desorption drying;
(iii) the thermophysical properties are constant but different for different regions;
(iv) the temperature T_d at the desorption interface is a known constant but the concentrations of vapour, C_d, and air, C_{ad}, at the interface are unknown constants, which should be determined in the solving process;

the desorption drying can be formulated as follows.

For the desorption mushy region:

$$\frac{\partial T_1}{\partial t} = k_1 \frac{\partial^2 T_1}{\partial x^2} + \rho H_p \frac{\partial f}{\partial t} \tag{12a}$$

$$\frac{\partial C_1}{\partial t} = \alpha_{m1} \frac{\partial^2 C_1}{\partial x^2} - \rho \frac{\partial f}{\partial t} \tag{12b}$$

$$\frac{\partial C_{a1}}{\partial t} = \alpha_{m1} \frac{\partial^2 C_{a1}}{\partial x^2} \tag{12c}$$

For the dried region:

$$\frac{\partial T_2}{\partial t} = k_2 \frac{\partial^2 T_2}{\partial x^2} \tag{12d}$$

$$\frac{\partial C_2}{\partial t} = \alpha_{m2} \frac{\partial^2 C_2}{\partial x^2} \tag{12e}$$

$$\frac{\alpha C_{a2}}{\partial t} = \alpha_{ma2} \frac{\partial^2 C_{a2}}{\partial x^2} \tag{12f}$$

The corresponding initial and boundary conditions are

$$t = 0, \quad T_1 = T_0, \quad C_1 = C_0, \quad C_{a1} = C_{a0}, \quad f = f_0 \tag{13a}$$

$$x = 0, \quad T_2 = T_s, \quad C_2 = C_s, \quad C_{a2} = C_{as}, \quad f = f_d \tag{13b}$$

$$x = \infty, \quad T_1 = T_0, \quad C_1 = C_0, \quad C_{a1} = C_{a0}, \quad f = f_0 \tag{13c}$$

The matching conditions at the desorption interface are

$$T_1 = T_2 = T_d \tag{14a}$$

$$C_1 = C_2 = C_d \tag{14b}$$

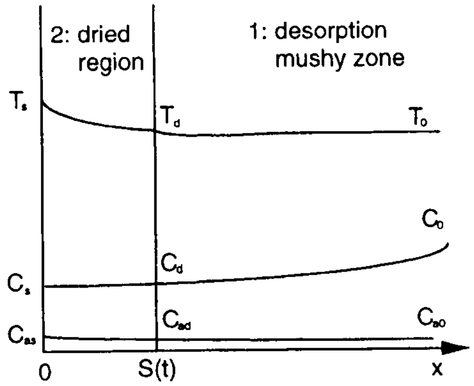

Figure 1. Physical model

$$C_{a1} = C_{a2} = C_{ad} \tag{14c}$$

$$f = f_d \tag{14d}$$

The energy and mass balance at the interface yield

$$-k_2 \frac{\partial T_2}{\partial x}\bigg|_{x=s} + k_1 \frac{\partial T_1}{\partial_x}\bigg|_{x=s} = 0 \tag{15a}$$

$$\alpha_{m2} \frac{\partial C_2}{\partial x}\bigg|_{x=s} - \alpha_{m1} \frac{\partial C_1}{\partial x}\bigg|_{x=s} = 0 \tag{15b}$$

$$-\alpha_{ma2} \frac{\partial C_{a2}}{\partial x}\bigg|_{x=s} + \alpha_{ma1} \frac{\partial C_{a1}}{\partial x}\bigg|_{x=s} = 0 \tag{15c}$$

3.2 Solution

As in previous work (Peng and Chen, 1994), we only treat the special case where the volumetric fraction of sorbed moisture is a linear function of temperature:

$$f = f_0 - \frac{f_0 - f_d}{T_d - T_0}(T_1 - T_0) \tag{16}$$

Equations (12)–(16) can be solved by the method stated in our previous work (Peng and Chen, 1994). By using the dimensionless parameters defined in nomenclature, the exact solutions can be obtained as follows:

$$\bar{T}_1(\eta) = \bar{T}_0 + \left(\bar{T}_d - \bar{T}_0\right)\frac{\operatorname{erfc}\,(\eta\,\alpha_{21}^{1/2})}{\operatorname{erfc}\,(\lambda\,\alpha_{21}^{1/2})} \tag{17a}$$

$$\bar{C}_1(\eta) = \bar{C}_0 - \Delta\bar{T}_0 + \left[\left(\bar{C}_d - \bar{C}_0\right) - \Delta\left(\bar{T}_d - \bar{T}_0\right)\right]\frac{\operatorname{erfc}\left(\eta[\alpha_{21}/Lu_1]^{1/2}\right)}{\operatorname{erfc}\left(\lambda[\alpha_{21}/Lu_1]^{1/2}\right)} \tag{17b}$$

$$+\Delta\left[\bar{T}_0 + \left(\bar{T}_d - \bar{T}_0\right)\frac{\operatorname{erfc}\,(\eta\,\alpha_{21}^{1/2})}{\operatorname{erfc}\,(\lambda\,\alpha_{21}^{1/2})}\right]$$

$$\bar{C}_{a1}(\eta) = \bar{C}_{a0} + \left(\bar{C}_{ad} - \bar{C}_{a0}\right)\frac{\operatorname{erfc}\left(\eta[\alpha_{21}/Lu_{a1}]^{1/2}\right)}{\operatorname{erfc}\left(\lambda[\alpha_{21}/Lu_{a1}]^{1/2}\right)} \tag{17c}$$

$$\bar{T}_2(\eta) = \bar{T}_s + \left(\bar{T}_d - \bar{T}_s\right) \frac{\operatorname{erf}(\eta)}{\operatorname{erf}(\lambda)} \tag{17d}$$

$$\bar{C}_2(\eta) = \bar{C}_s + \left(\bar{C}_d - \bar{C}_s\right) \frac{\operatorname{erf}\left(\eta\,[Lu_2]^{-1/2}\right)}{\operatorname{erf}\left(\lambda[Lu_2]^{-1/2}\right)} \tag{17e}$$

$$\bar{C}_{a2}(\eta) = \bar{C}_{as} + \left(\bar{C}_{ad} - \bar{C}_{as}\right) \frac{\operatorname{erf}\left(\eta\,[Lu_{a2}]^{-1/2}\right)}{\operatorname{erf}\left(\lambda\,[Lu_{a2}]^{-1/2}\right)} \tag{17f}$$

The energy and mass balance equations become

$$kT \frac{\exp\{-\lambda^2\}}{\operatorname{erf}(\lambda)} - \alpha_{21}^{1/2} \frac{\exp\{-\lambda^2 \alpha_{21}\}}{\operatorname{erfc}(\lambda \alpha_{21}^{1/2})} = 0 \tag{18a}$$

$$\begin{aligned} &\left[\frac{\alpha_{21}\alpha_{m21}}{Lu_1}\right]^{1/2} \left(\bar{C}_d - \bar{C}_s\right) \frac{\exp\{-\lambda^2\alpha_{21}/\alpha_{m21}Lu_1\}}{\operatorname{erf}\left(\lambda[\alpha_{21}/\alpha_{m21}Lu_1]^{1/2}\right)} \\ &+ \left[\frac{\alpha_{21}}{Lu_1}\right]^{1/2} \left[\left(\bar{C}_d - \bar{C}_0\right) - \Delta\left(\bar{T}_d - \bar{T}_0\right)\right] \frac{\exp\{-\lambda^2\alpha_{21}/Lu_1\}}{\operatorname{erfc}\left(\lambda\,[\alpha_{21}/Lu_1]^{1/2}\right)} \\ &+ \alpha_{21}^{1/2}\Delta\left(\bar{T}_d - \bar{T}_0\right) \frac{\exp\{-\lambda^2\alpha_{21}\}}{\operatorname{erfc}(\lambda\ \alpha_{21}^{1/2})} = 0 \end{aligned} \tag{18b}$$

$$\alpha_{ma21}^{1/2}\ \frac{\bar{C}_{as} - \bar{C}_{ad}}{\bar{C}_{ad} - \bar{C}_{a0}}\ \frac{\exp\{-\lambda^2/Lu_{a2}\}}{\operatorname{erf}\left(\lambda\,[Lu_{a2}]^{-1/2}\right)} - \frac{\exp\{-\lambda^2\alpha_{21}/Lu_{a1}\}}{\operatorname{erfc}\left(\lambda\,[\alpha_{21}/Lu_{a1}]^{1/2}\right)} = 0 \tag{18c}$$

The desorption interface position parameter λ can be determined by equation (18a), and the interface concentrations of vapour and air can be determined by equations (18b) and (18c), respectively. As soon as the interface parameters are determined, equations (17) easily result in the exact solution of the desorption drying of a porous half-space.

3.3 Analysis and discussion

For desorption drying, the most important parameter is desorption rate, which is reflected by the desorption interface parameter, λ. A larger λ means faster desorption. The additional interest may be in vapour concentration distribution, which is reflected by the vapour concentration at the desorption interface.

The analysis of air concentration distribution is ignored, because it is of less importance for our problem. Desorption occurs in a vacuum environment, and no air source exists in the process, but the vapour is continuously produced by desorption of sorbed moisture, so that the air is completely exhausted shortly after the beginning, and the vapour diffusion can be considered as mutual diffusion in the limiting case with a negligibly small amount of air.

According to the experimental results of Mellor (1978, pp. 155–159), the air in the porous sublimed layer is exhausted soon after the beginning of sublimation drying, so that the partial pressure of vapour at the surface of the porous sublimed layer is nearly equal to the total vacuum pressure. Although for desorption drying no similar experiment can be found in the literature, however, we can infer that the conclusion holds even for desorption drying because it usually proceeds in a higher vacuum environment so that the amount of air in the porous medium is far less than that of vapour and the diffusion is the mutual diffusion in the limiting case. The limiting mutual diffusion should not be confused with the self-diffusion of vapour in a porous medium. According to Sherwood *et al.* (1975, p. 14), the mutual diffusivity in the limiting case is not equal to the self-diffusivity.

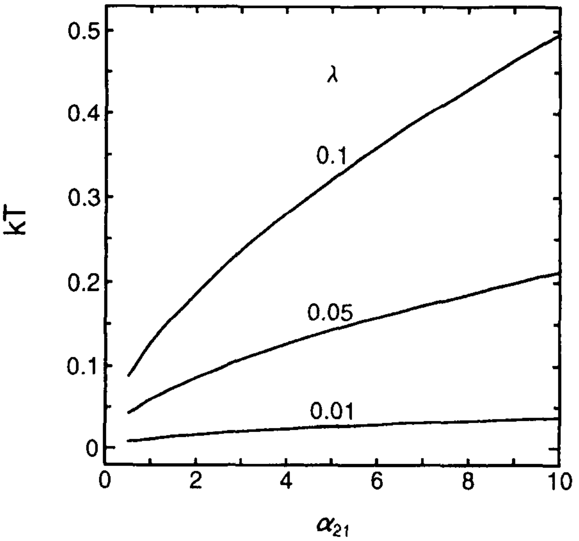

Figure 2. The effects of α_{21} and kT on the rates of desorption

For the exact solutions obtained, we have made a numerical analysis to demonstrate the effect of various parameters on the desorption rate and vapour concentration at the desorption interface; the results are shown in Figures 2 and 3, respectively. On the figures, only the parameters whose values are different from the reference values are indicated. The selected reference values include: $\lambda = 0{\cdot}01$, $Lu_1 = 10$, $\alpha_{21} = 3$, $\Delta = -3$, $\overline{T}_d = 1{\cdot}07$, $\overline{T}_0 = 1$, $\overline{C}_s = 0{\cdot}01$, $\overline{C}_0 = 0{\cdot}1$, $f_0 = 0{\cdot}05$, $f_d = 0{\cdot}03$. The calculation results apply to the desorption drying of the porous medium of sand with moisture.

Figure 2 shows the effects of the ratio of thermal diffusivities, α_{21}, and the ratio of the steady heat fluxes, kT, between the dried region and the desorption mushy zone, on the rate of desorption. A larger α_{21} always results in slower desorption, but a larger kT results in faster desorption. A large α_{21} means a large sorbed moisture content (if the thermophysical properties and the temperatures remain unchanged),

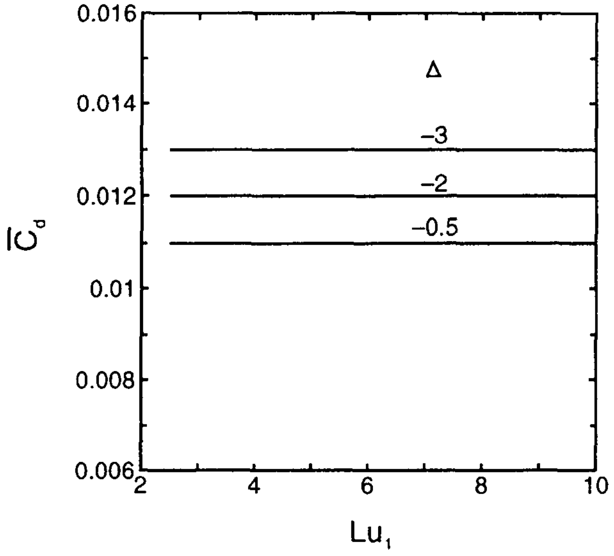

Figure 3. The effects of Δ and Lu_1 on the vapour concentration

which lowers the desorption rates; a large kT means a large temperature difference across the porous medium, which raises the desorption rates.

Figure 3 shows the effect of the desorption parameters, Δ, and the Luikov number, Lu_1, on the interface vapour concentration $\bar{C}_d$. A larger Δ, which means a smaller amount of sorbed moisture content, results in a smaller vapour concentration. A larger Lu_1 doesn't affect the vapour concentration very much.

4. CONCLUSION

A novel desorption drying theory has been developed, and the equations derived to describe transient heat and mass transfer in desorption drying.

The present theory at least improves upon that of King in following three aspects: (i) it is directly related to the real transport mechanism, in contrast to King's indirect equation; (ii) it is transient in both heat and mass transfer; (iii) it is even applicable to the last phase of desorption drying where King's theory breaks down.

By using the desorption mushy zone model together with the present theory, an exact solution has been obtained for heat and mass transfer with desorption in a porous half-space, with results analysed.

The results indicate that the rates of desorption are lowered with an increase in α_{21}, but are raised with an increase in kT. The vapour concentration diminishes with an increase of Δ.

NOMENCLATURE

c = specific heat (J kg^{-1} K^{-1})
C = M_v/V_f, concentration of moisture (kg m^{-3})
C_a = M_a/V_f, concentration of air (kg m^{-3})
C_s = M_s/V_f, concentration of solid skeleton, or surface vapour concentration (kg m^{-3})
$\bar{C}$ = C/C_3, non-dimensional concentration of vapour
$\bar{C}_a$ = C_a/C_3, non-dimensional concentration of air
D_{eff} = effective 'diffusivity' of sorbed moisture (m^2 s^{-1})
f = V_v/V_f, adsorption water volume fraction
h = enthalpy (J kg^{-1})
H_p = desorption heat (J kg^{-1})
j = mass flux (kg m^{-2} s^{-1})
k = effective thermal conductivity (W mK^{-1})
kT = $k_2(\bar{T}_s - \bar{T}_d)/[k_1(\bar{T}_d - \bar{T}_0)]$
Lu = α_m/α, Luikov number of vapour diffusion
Lu_a = α_{ma}/α, Luikov number of air diffusion
M = weight (kg)
q = heat flux (W m^{-2})
$S(t)$ = position of second desorption front (m)
t = time (s)
T = temperature (K)
$\bar{T}$ = T/T_3, non-dimensional temperature
V = volume (m^3)
x = space coordinate (m)

Greek symbols

α = thermal diffusivity (m^2 s^{-1})
α_m = diffusivity of vapour (m^2 s^{-1})

α_{ma} = diffusivity of air ($m^2\ s^{-1}$)

α_{21} $= \alpha_2/\alpha_1^*$

α_1^* $= \left\{ \dfrac{1}{\dfrac{1}{\alpha_1} + \dfrac{\rho H_p}{k_1}\,\dfrac{f_0 - f_d}{T_d - T_0}} \right\}^{-1}$, effective thermal diffusivity in desorption mushy zone ($m^2\ s^{-1}$)

φ_1 $= \rho\alpha_1^* \dfrac{f_0 - f_d}{T_d - T_0}$, desorption parameter ($kg\ m^{-1}\ s^{-1}\ K^{-1}$)

ρ = density of sorbed water ($kg\ m^{-3}$)

λ $= S(t)/2[\alpha_2 t]^{1/2}$, position parameters at the second desorption front

η $= x/2[\alpha_2 t]^{1/2}$

β $= \dfrac{\varphi_1}{\alpha_{m1} - \alpha_1^*}$ ($kg\ m^{-3}\ K^{-1}$)

Δ $= -\dfrac{\beta T_3}{C_3}$

Subscripts

0 = initial
1 = desorption mushy region
2 = dried region
3 = triple point
a = air
d = secondary desorption front
f = framework
s = solid skeleton; surface
v = vapour
w = sorbed water, or called sorbed moisture

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