# DRYING OF HYGROSCOPIC CAPILLARY POROUS SOLIDS  $-A$  THEORETICAL APPROACH

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**Abstract---A** mathematical model of drying is presented whereby capillary flow of liquid and vapour diffusion as well as heat transfer through sohds are taken into account simultaneously. The Clausius-Ciapeyron's equation or the sorptional isotherm is used as coupling equation depending upon the stage of drying. The results in the form of traditional rate of drying curves are obtained and discussed with various internal and external physical parameters



vapour



# Subscripts



# **1. INTRODUCTION**

**THE DRYING** of solids is usually taken to mean the removal of a liquid from a moist solid by evaporation [1]. The heat required for evaporation has to be supplied to the material, therefore heat and mass transfer occur simultaneously. Usually the evaporated moisture is carried away by an external drying media (often air) circulated above the drying solid. Since the general drying behaviour under constant external conditions and the definitions of the various drying periods are well known [2, 31, only a brief summary of the theoretical approaches suggested in the past will be presented.

Neglecting the effect of heat transfer completely, the early theoretical studies (1936 1937) assumed that moisture was transferred either by diffusion [4-S] or by capillary action [9-11]. This oversimplification, applicable under very specific circumstances, had not only led to wrong predictions in some general situations but, in other cases, to misinterpretation of experimental results. Consequently, more elaborate models were proposed, and, at present, basically two approaches are widely quoted in literature.

Krischer's model  $[12-15]$  was based on the assumption that, during the drying, some combination of capillary flow of liquid and diffusion of vapour controlled the internal moisture transfer. Luikov's approach [16-19], on the other hand, was an attempt to establish a theoretical model by applying the methods of the thermodynamics of irreversible processes to the transport phenomena during the drying process. Both postulated that at the surface of the drying material the corresponding equilibrium values of the dependent variables were reached instantaneously at the beginning of the drying process. This assumption of surface boundary conditions of the first kind, together with the application of the sorptional isotherm for the whole range of liquid (moisture) contents are probably the main difticulties encountered in Krischer's model in predicting drying rate curves. Luikov, assumed that the "moisture diffusivity" and the "specific moisture capacity" are constant, which leads to an apparent mathematical paradox if his model is extended to the non-isothermal drying.

In summary, despite the fact that efforts have been made in the past to develop a theoretical model for the drying process, the experimental approach still remains the preferred route in studying the problem related to drying [l]. It is, therefore, the purpose of this paper to present a new approach to this relatively complex problem of simultaneous heat-and mass-transfer processes. The model to be described is based on Krischer's basic theories. However, modifications and extensions are made **such** that the dynamic drying behaviour, i.e. the drying rate curve can be predicted once the basic physical parameters of the system are known.

# **2. THE BASIC THEORY**

The physical model underlying the mathematical statement can be described as follows:

(1) The total internal moisture transfer during drying can be divided into two parts (Fig. 1): the capillary flow of liquid due to a gradient in liquid content, and the diffusion of vapour through "empty" pores due to a gradient in



Fig. 1. Sketch, illustrating the theoretical model.

partial vapour pressure. The internal heat transfer is governed by the heat conduction through the solid skeleton and the latent heat of phase changes.

(2) Inside the moist solid, the rate of phase change is much faster than either the rates of heat and mass transfer. Thus, liquid content, partial vapour pressure and temperature are in equilibrium at any location within the drying material.

(3) Consequently, for liquid contents larger than the maximum sorptional moisture content, the partial vapour pressure is equal to its saturation value. For the sorptional region of liquid content, the partial vapour pressure is controlled by the sorptional isotherm.

(4) The external heat and mass transfer is assumed to be proportional, respectively, to the temperature and the partial vapour pressure difference between the surface of the drying solid and the external drying media.

If the liquid content, u, is defined by  $u = v_I /$  $v<sub>s</sub>$  and assuming that Fick's law is applicable to both mass-transfer mechanisms, the liquid flux,  $\dot{m}_L$ , towards the surface of the drying material (Fig. 1) is

$$
\dot{m}_L = -K_L \rho_L \frac{\partial u}{\partial x} \tag{1}
$$

With the supposition that for small temperature changes the partial vapour pressure gradient is proportional to the vapour density gradient, i.e.

$$
\frac{1}{R_v T} \frac{\partial p_v}{\partial x} = \frac{\partial}{\partial x} \left[ \frac{p_v}{R_v T} \right] = \frac{\partial \rho_v}{\partial x}
$$
 (2)

the vapour flux,  $\dot{m}_v$ , towards the surface can be represented by

$$
\dot{m}_v = -K_v(\varepsilon - u) \frac{\partial \rho_v}{\partial x} \tag{3}
$$

where  $K_v$  is the vapour diffusivity of the solid.

If  $\dot{m}_{\alpha}$  denotes the rate of evaporation inside the drying material, and both  $K_L$  and  $K_v$  are assumed to be constant, a mass balance over a small volume element of unit cross sectional area and thickness, dx, leads to the differential equation for the liquid transfer,

$$
K_{L}\rho_{L}\frac{\partial^{2} u}{\partial x^{2}} - \dot{m}_{\infty} = \rho_{L}\frac{\partial u}{\partial t}
$$
 (4)

and the differential equation of vapour transfer

$$
K_{\nu}\frac{\partial}{\partial x}\left[(\varepsilon-u)\frac{\partial\rho_{\nu}}{\partial x}\right]+\dot{m}_{\infty}=\frac{\partial}{\partial t}\left[(\varepsilon-u)\rho_{\nu}\right].
$$
 (5)

Differentiating equation (5) by parts and eliminating  $\dot{m}_{e}$ , the total moisture transfer is obtained in the form of

$$
K_{L}\rho_{L}\frac{\partial^{2} u}{\partial x^{2}} + K_{v}\left[ (\varepsilon - u)\frac{\partial^{2} \rho_{v}}{\partial x^{2}} - \frac{\partial u}{\partial x} \frac{\partial \rho_{v}}{\partial x} \right]
$$

$$
= (\rho_{L} - \rho_{v})\frac{\partial u}{\partial t} + (\varepsilon - u)\frac{\partial \rho_{v}}{\partial t}.
$$
(6)

Assuming that heat is transferred only by conduction through the solid skeleton, i.e. the heat flux,  $\dot{q} = -K_T \frac{\partial T}{\partial x}$ , and realizing that the rate of latent heat required for or liberated by phase changes is  $\dot{q}_{\alpha} = r_v \dot{m}_{\alpha}$  a heat balance leads to

$$
\alpha \frac{\partial^2 T}{\partial x^2} + \frac{r_v}{\rho_s c_s} \left\{ K_v \left[ (\varepsilon - u) \frac{\partial^2 \rho_v}{\partial x^2} - \frac{\partial u}{\partial x} \frac{\partial \rho_v}{\partial x} \right] - (\varepsilon - u) \frac{\partial \rho_v}{\partial t} + \rho_v \frac{\partial u}{\partial t} \right\} = \frac{\partial T}{\partial t} \qquad (7)
$$

where  $K_L$  is the liquid conductivity in the solid. as differential equation for the heat transfer.

With the rate of drying,  $\dot{r}_D = K_c$  ( $\rho_v|_{x=0}$  - $\rho_{\text{val}}$ , the boundary condition at  $x = 0$  for equation (6) becomes

$$
K_L \rho_L \frac{\partial u}{\partial x} + K_v (\varepsilon - u) \frac{\partial \rho_v}{\partial x}
$$
  
=  $K_c (\rho_v - \rho_{va})$ , at  $x = 0$  (8)

which is the mathematical statement of the fact that the sum of liquid and vapour flux to the surface must be equal to the vapour flux away from the surface into the external drying media. Similarly, the corresponding boundary condition for equation (7) becomes

$$
h(T_a - T) = r_v K_L \rho_L \frac{\partial u}{\partial x} - K_T \frac{\partial T}{\partial x}, \text{ at } x = 0 \quad (9)
$$

where the term  $r_v K_L \rho_L \partial u / \partial x |_{x=0}$  denotes the amount of heat required to evaporate the liquid flux at the surface.

For the boundary conditions at  $x = l$  it is assumed that no mass and heat is transferred across that surface of the drying material. Thus,

$$
K_L \rho_L \frac{\partial u}{\partial x} = K_v (\varepsilon - u) \frac{\partial \rho_v}{\partial x} = 0, \text{ at } x = l. \quad (10)
$$

$$
\frac{\partial T}{\partial x} = 0, \text{ at } x = l. \quad (11)
$$

As stated previously, this approach, in contrast to Krischer's model, employs two coupling equations between the three dependent variables  $u, \rho$ , and T. These two equations are:

(1) The Clausius-Clapeyron's equation in the form of

$$
\rho_{vm} = \frac{1}{R_v T}
$$
  
exp  $\left(58.7395 - \frac{6847.96}{T} - 5.262 \ln T \right)^*$  (12)

where  $R<sub>v</sub>$  is the gas constant of the vapour in  $m^2$ <sup>o</sup>K. \_. -\_ ~~~-.- -

(2) The equation of the sorptional isotherm of the system which, in general, can be represented by

$$
\rho_v = \psi_1(u, T). \tag{13a}
$$

To separate the regions of applicability of equations (12) and (13a), it is reasonable to assume the maximum sorptional liquid content,  $u_{\rm sm}$ , to be a function of temperature only, i.e.

$$
u_{\rm sm} = \psi_2(T). \tag{13b}
$$

At this stage, it should be pointed out that the explicit forms of equations (13) are, in general, determined experimentally.

Now it is obvious that equations  $(6)$ – $(13)$  will completely describe the drying process for any conditions imposed by the external drying medium. It must, however, be remembered that for liquid contents larger than  $u_{\rm sm}$  the vapour density has to be calculated from equation (12) while for all other values of  $u$  the vapour density is obtained from equation (13).

## 3. NUMERICAL SOLUTIONS AND RESULTS

To facilitate the numerical solutions of these differential equations, the following dimensionless variables are introduced :

(a) Independent variables *:* 

$$
\xi = \frac{x}{l} \tag{14a}
$$

$$
N_{F0} = \frac{\alpha t}{l^2} \tag{14b}
$$

(b) Dependent variables

$$
U = \frac{u - u_e}{u_0 - u_e} = \frac{u - u_e}{\Delta u}
$$
 (14c)

$$
V = \frac{\rho_v - \rho_{v_e}}{\rho_{v_0} - \rho_{v_e}} = \frac{\rho_v - \rho_{v_e}}{\Delta \rho_v} \qquad (14d)
$$

$$
\theta = \frac{T_e - T}{T_e - T_0} = \frac{T_e - T}{\Delta T}.
$$
 (14e)

<sup>\*</sup> The constants of this equation can bc obtained when substituting the thermodynamic properties of water in mgs-system into the theoretical equation. For the derivation of equation (12) see [20].

Substitution of these variables into equations  $(6)$ – $(11)$  leads to the dimensionless differential equations :

$$
N_{\mu\nu_{L}} \frac{\rho_{L}}{\rho_{v}} \frac{\partial^{2} U}{\partial \xi^{2}} + N_{\mu\nu_{v}} \left[ (U_{\epsilon} - U) \frac{\partial^{2} V}{\partial \xi^{2}} - \frac{\partial U}{\partial \xi} \frac{\partial V}{\partial \xi} \right]
$$
  
=  $\left( \frac{\rho_{L} - \rho_{v_{e}}}{\Delta \rho_{v}} - V \right) \frac{\partial U}{\partial N_{F0}} + (U_{\epsilon} - U) \frac{\partial V}{\partial N_{F0}} \quad (15)$ 

$$
\frac{\partial^2 \theta}{\partial \xi^2} - N_{Ko} \frac{\Delta \rho_v}{\rho_s} N_{Lu_v} \left[ (U - U) \frac{\partial^2 V}{\partial \xi^2} - \frac{\partial U}{\partial \xi} \frac{\partial V}{\partial \xi} \right]
$$

$$
- (U_{\varepsilon} - U) \frac{\partial V}{\partial N_{Fo}} + \left( V + \frac{\rho_{v_e}}{\Delta \rho_v} \right) \frac{\partial U}{\partial N_{Fo}} \}
$$

 $=\frac{\partial \theta}{\partial x}$  $\partial {N}_{Fo}$ 

and the dimensionless boundary conditions become

$$
N^*_{Bi_m}(V - V_a) = N_{L \mathbf{u}_L} \frac{\Delta u \rho_L}{\Delta \rho_v} \frac{\partial U}{\partial \xi}
$$
 and  
+  $N_{L \mathbf{u}_v} \Delta u (U_{\varepsilon} - U) \frac{\partial V}{\partial \xi}$ , at  $\xi = 0$  (17a)

$$
\frac{\partial U}{\partial \xi} = 0, \text{ at } \xi = 1 \tag{17b}
$$

$$
N_{\mathcal{B}i_{\mathsf{h}}}\theta = N_{\mathcal{K}o}N_{\mathcal{L}u_{\mathcal{L}}}\frac{\rho_{\mathcal{L}}}{\rho_{\mathsf{s}}}\frac{\partial U}{\partial \xi} + \frac{\partial \theta}{\partial \xi}, \text{at } \xi = 0 \quad (17c)
$$

$$
\frac{\partial \theta}{\partial \xi} = 0, \text{ at } \xi = 1. \tag{17d}
$$

The coupling equations (12) and (13) become

$$
V_m = \left\{ \frac{1}{R_v(\Delta T \theta + T_e)} \exp \left[ 58.7395 - \frac{6847.96}{\Delta T \theta + T_e} - 5.262 \ln (\Delta T \theta + T_e) \right] - \rho_{v_e} \right\} / \Delta \rho_v \qquad (18)
$$

$$
V = \psi_3(U, \theta); \tag{19a}
$$

$$
U_{s_m} = \psi_4(\theta). \tag{19b}
$$

Finally, the rate of drying in dimensionless notation can be written as

$$
\dot{R}_D = V|_{\xi=0} - V_a, \text{ where } \dot{R}_D = \dot{r}_D/(K_c \cdot \Delta \rho_v).
$$

To summarize, a list of the dimensionless groups is given in Table 1 with their respective physical interpretations.

For the numerical solutions of the described model, the general relationships as stated in equation (19) must be replaced by some mathematical equations. Since, in general, the sorptional isotherms show an S-shaped characteristic (in a  $\rho_v/\rho_{v_{sm}}$  vs.  $u/u_{sm}$  plot), two approximations are chosen. Namely,

(16) 
$$
V = \left[ \frac{1}{2} \left\{ 1 + \sin \left[ \pi \left( \frac{\Delta u U + u_e}{\Delta u U_{s_m} + u_e} - \frac{1}{2} \right) \right] \right\} - \left( \Delta \rho_v V_m + \rho_{v_e} \right) - \rho_{v_e} \right] / \Delta \rho_v \qquad (20)
$$

$$
V = \left\{ C_1 \left[ \frac{1}{C_2} \left( \frac{\Delta u U + u_e}{\Delta u U_{s_m} + u_e} - C_3 \right) \right]^{\frac{1}{3}} \right\}
$$

$$
\times \left( \Delta \rho_v V_m + \rho_{v_e} \right) / \Delta \rho_v. \qquad (21)^*
$$

The sin-approximation (equation (20)) is selected for pure mathematical convenience while the 3rd order polynomial (equation (21)) could represent a first approximation of real sorptional isotherms.<sup>†</sup> The relationship for the maximum sorptional liquid content may be approximated by

$$
U_{s_m} = \left(S_1 + S_2 \frac{T_e - T}{\Delta T} - u_e\right) / \Delta u \,. \tag{22}
$$

The set of equations  $(15)-(17)$ , together with  $(18)$ ,  $(22)$ ,  $(20)$  or  $(21)$  were solved numerically using the method of finite differences. From the many possible difference schemes reported in

<sup>\*</sup> $C_1$ ,  $C_2$  and  $C_3$  can be correlated by two equations which express, let us say,  $C_1$  and  $C_2$  in terms of  $C_3$ .

t The sorption isotherm as given in Fig. 29 and Fig. 41 in [12] can be approximated by equation (4) to within  $± 10$  per cent.

Name	Symbol	Definition	Significance
Biot number (heat transfer)	$N_{Bi}$	hl $K_T$	Surface film resistance/Thermal internal resistance (External parameter)
Biot number (mass transfer modified)	$N_{Bi_m}^*$	$K_c$ l $\alpha$	Mass-transfer rate at interface/ Heat-transfer rate in interior of solids (External parameter)
Kossovich number	$N_{K_{\rm v}}$	$r_v, \Delta u$ $\overline{C \Delta T}$	Heat used for evaporation/Heat used in raising temperature of solid
Luikov number (liquid transfer)	$N_{L_{u_L}}$	$K_L$ $\alpha$	Rate of liquid transfer/Rate of heat transfer (Internal parameter)
Luikov number (vapour transfer)	$N_{t}$	$K_v$ $\boldsymbol{\chi}$	Rate of vapour transfer/Rate of heat transfer (Internal parameter)
	$\dot{R}_D$	$\frac{\dot{r}_D}{K_c(\rho_{v_0}-\rho_{v_0})}$	Actual rate of drying/Initial rate of drying

Table 1. Dimensionless parameters in drying processes

literature  $[21, 22]$ , the implicit scheme suggested by Crank and Nicolson [23] was employed. The resulting system of simultaneous non-linear algebraic equations were solved by an iterative procedure similar to the method of successive overrelaxation. The convergence of the method was assured by decreasing the time step while keeping the grid ratio constant until a change in the drying rate curve could no longer be observed.

The following range of parameters were investigated:

$$
0.02 \leq N_{Lu_L} \leq 0.4
$$
  
\n
$$
5 \leq N_{Lu_v} \leq 500
$$
  
\n
$$
1500 \leq N_{B_{in}}^* \leq 3000
$$
  
\n
$$
0.083 \leq N_{B_{in}} \leq 2.
$$

The drying rate curves shown in Figs. 2-5 were calculated using equation (20) as sorptional isotherm, and  $S_1 = 0.14$ ,  $S_2 = -0.0005$  as constants in equation (22). Figure 6 shows drying rate curves obtained for different sorptional



Fig. 2. Drying rate curves— $\hat{R}_D$  vs.  $N_{F_0}$ . Parameter: Internal liquid transfer,  $N_{Lu_{L}}$ .



Fig. 3. Drying rate curves— $\vec{R}_D$  vs.  $N_{F_0}$ . Parameter: Internal vapour transfer,  $N_{L u_v}$ .





FIG. 4. Drying rate curves— $\mathring{R}_D$  vs.  $N_{f\alpha}$  (curves (a)), and  $\dot{r}_D$  vs.  $N_{f\alpha}$  (curves (b)). Parameter: External mass transfer,  $\tilde{N}_{B_{im}}^{*}$  (curves having the same arabic number correspond).

Fig. 6. Drying rate curves— $\vec{R}_D$  vs.  $N_{F_o}$ . Parameter: Sorption isotherm.



Fig. 5. Drying rate curves— $\vec{R}_D$  vs.  $N_{F_0}$ . Parameter: External heat transfer,  $N_{Bi_h}$ .



FIG. 7. Dimensionless liquid content,  $U$ , and temperature,  $T$ , at the surface as a function of the drying time,  $N_{F_0}$  (V indicates the beginning of the falling rate period).

isotherms. The variation of temperature and liquid content at the surface of the drying material with respect to the drying time is shown in Fig. 7.

## 4. DISCUSSION AND CONCLUSION

All calculated drying rate curves show the characteristics which are well known from experimental investigations. After an initial increase or decrease of the rate of drying (RD), the drying process enters the constant rate period (CRP). This initial change of the RD is caused by a variation of the surface temperature (Fig. 7) which in turn results in a change of vapour density. It can be seen from Fig. 2 to Fig. 5 that this interval of the drying process is solely determined by external drying conditions.

Referring to curves (a) in Fig. 4 it must be

remembered that the dimensionless rate of drying,  $\dot{R}_b$ , has been defined as the ratio of the actual RD to the initial RD, the latter being dependent upon  $K_c$ . Thus, curves (a) in Fig. 4 indicate that for large  $N_{B_{im}}^*$  the actual RD decreases compared to the initial RD. In plotting the actual RD vs.  $N_{ro}$ (curves(b)) one immediately recognizes that the actual RD increases with increasing mass transfer coefficient,  $K_c$ . The effect of the external heat transfer on this initial interval can be seen from Fig. 5. For large values of  $N_{Bis}$ , the heat supply to the surface from the external drying media is larger than the amount of latent heat required for evaporation at the surface (curve (a) in Fig. 5). Consequently, the surface temperature rises resulting in an increase of the vapour density, and thus, increasing the RD. **The** opposite is true for small values of  $N_{Bi_h}$ , where additional heat has to be transported from within the drying material to balance the latent heat requirement at the surface.

At the beginning of the CRP, the surface temperature will assume a value at or close to the wet bulb temperature of the drying media, and a quasi-thermodynamic equilibrium prevails at the surface during the CRP (Fig. 7). It is, therefore, not surprising that the value of the RD for this period is solely determined by external conditions (Fig. 4 and Fig. 5). The duration of the CRP, however, is almost entirely controlled by  $N_{Lu_L}$ , i.e. the internal liquid transfer (Fig. 2). Since with increasing  $N_{Lur}$ , the duration of the CRP also increases, one could conclude that the CRP last as long as the liquid flux to the surface from within the solid can compensate for the rate of evaporation at the surface. However, from Fig. 7 it can be seen that the CRP ends once the liquid content at the surface drops below its maximum sorptional value. Now, since during the CRP, the RD is controlled by the external conditions, the liquid content gradient at the surface has to be larger for small liquid conductivities of the solid material. Consequently, the maximum sorptional liquid content is reached faster for

small values of the liquid conductivity. Therefore, solids with smaller liquid conductivities have a shorter CRP.

Once the drying process has entered the falling rate period (FRP), the external conditions become relatively unimportant compared to the internal parameters. This can be deduced from the fact that the drying rate curves for the FRD in Fig. 4 and Fig. 5 are parallel to each other while there is a marked difference in the corresponding regions of Fig. 2 and Fig. 3. The vapour transfer which up to this point did not affect the drying behaviour has an increasing influence as the drying proceeds into the FRP (Fig. 3). Figure 6 shows drying rate curves with the sorptional isotherm as parameter. From these curves it is obvious that the characteristic of the drying behaviour during the FRP is, to a large extent, controlled by the sorptional isotherm of the system.

In conclusion, it can be said that the approach to the drying of porous solids presented here, leads to theoretical predictions which can reasonably be explained from the physical point of view. Clearly, the final verification can only be reached by systematic experimental investigations which, at present, are being carried out.

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# **302** D. BERGER and D. C. T. PEI

## SECHAGE DE SOLIDES MICRO-POREUX HYGROSCOPIQUES- UNE APPROCHE **THEORIOUE**

Résumé- On présente un modèle mathématique de séchage dans lequel sont pris en compte simultanément l'écoulement capillaire du liquide, la diffusion de vapeur, aussi bien que le transfert thermique dans les solides. L'équation de Clausius-Clapeyron ou l'isotherme de sorption est utilisée comme une équation de couplage dépendant du degré de séchage. Les résultats, sous forme de courbes de taux de séchage. sont obtenus et discutes a partir des nombreux parametres physiques internes et externes.

#### TROCKNEN VON HYGROSKOPISCHEN, KAPILLARPORÖSEN KÖRPERN-EINE THEORETISCHE NAHERUNG

Zusammenfassung-Es wird ein mathematisches Modell für die Trocknung angegeben, bei dem sowohl kapillare Flüssigkeitsströmung und Dampfdiffusion als auch Wärmeleitung durch feste Körper gleichzeitig berücksichtigt werden. Als Kopplungsgleichung wurde die Gleichung von Clausius-Clapeyron oder die Sorptionalisotherme benutzt, die vom Trocknungsgrad abhangt. Die Ergebnisse werden in der Form des traditionellen Anteils der Trocknungskurven erhalten und diskutiert mit verschiedenen inneren und ausseren physikalischen Parametern.

#### СУШКА ГИГРОСКОПИЧЕСКИХ КАПИЛЛЯРНО-ПОРИСТЫХ МАТЕРИАЛОВ. ТЕОРЕТИЧЕСКИЙ АНАЛИЗ

Аннотация—Представлена математическая модель процесса сушки, одновременно чуитывающая капиллярное течение, диффузию нара и теплообмен в твердом теле. В зависимости от нернода сушки в качестве связующего уравнения использовалось манение Клаузиуса-Кланейрона или изотерма сорбции. Полученные результаты обработаны графически в виде обычных кривых скорости сушки для различных внутренних и внешних физических параметров.