

PORE NETWORK MODELING OF CONVECTIVE DRYING

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ABSTRACT

This paper presents a coupled pore-network model that accounts for the isothermal evaporation of a volatile liquid within a porous medium, mass transfer of the liquid vapors by diffusion and flow through liquid films within the porous medium, and convective mass transfer through the mass boundary layer over the surface of the porous medium. We study the effect of the thickness of the mass boundary layer and the Peclet number of the purge gas that flows over the surface of the porous medium on the drying curves. Our results show that the drying rate remains practically constant as long as the liquid films span along the pore network and their density at the surface is sufficiently high. This condition is satisfied when the thickness of the convective layer is greater than the average pore radius. The effects of the Peclet number and the velocity profile on the drying curves appear to be negligible. Our results explain previously reported experimental findings and provide a rigorous explanation of the constant-rate period.

1. INTRODUCTION

Drying of porous media is a process of significant scientific and applied interest. It involves several mechanisms at the pore scale that affect the macroscopic behaviour of the drying process. These mechanisms include phase change at the liquid-gas interfaces, mass and heat transfer by diffusion and convection, capillarity-induced flow through wetting liquid films and the receding of the liquid-gas interfaces under combined viscous, capillary and buoyancy forces.

In most typical applications, porous materials are subjected to a flow of a purge gas along the external porous surface, which can significantly enhance the recovery process and reduce drying times. The local mass transfer coefficient depends on the mass transfer conditions within the convective layer over the surface, as well, as on the liquid saturation at the surface.

For a realistic solution of the drying problem, mass transfer in the convective layer needs to be solved in conjunction with mass transfer within the porous medium.

This paper presents a coupled pore-network model that accomplishes such a solution. The model accounts for isothermal evaporation at the liquid-gas interface, mass transfer by diffusion and flow through liquid films within the porous medium and convective mass transfer through the mass boundary layer over the surface.

2. MODEL FORMULATION

We consider the isothermal drying of a porous medium saturated with a volatile single-component liquid (Fig. 1). All sides of the porous material, except one, are impermeable to fluid flow and mass transfer. The product surface (S) is exposed to a gas, which is flowing parallel to the surface with characteristic velocity U_∞ , purging the liquid vapors by convection and diffusion. A mass-transfer layer of thickness δ_m develops in the external gas, at the outer side of which the vapor pressure of the volatile component is zero. Periodic boundary conditions apply within the convective layer in the direction of the gas flow.

The mass boundary layer has been studied in great detail in the past (Bird et al., 1960; Leal, 1992). For boundary layer flow over a flat plate and a fixed surface concentration, δ_m is well approximated by

$$\delta_m = \delta \text{Sc}^{-1/3} \quad (1)$$

where δ is the momentum boundary-layer thickness, $\text{Sc} = \frac{D}{\nu}$ the Schmidt number and D is the gas-phase diffusivity. Typically, we have

$$\delta \approx 4.64X \text{Re}_X^{-1/2} \quad (2)$$

where $\text{Re}_X = \frac{U_\infty X}{\nu}$ is the local Reynolds number (at the downstream location X), U_∞ is the free-stream, characteristic gas velocity, and ν is the gas kinematic viscosity. In the context of the drying of a porous medium, the boundary layer will also be affected by two additional factors: surface S is permeable, and its surface concentration is non-uniform and variable. In this study we assume that the thickness of the mass boundary layer is independent of the Reynolds number Re_X . The effect of the later on δ_m will be addressed in a forthcoming publication.

Evaporation in a porous medium, uncoupled to external mass transfer has been discussed, in detail, in recent publications by Yiotis et al. (2001, 2003, 2004, 2005, and 2006). At any given time, one can identify three distinct regions inside the porous medium (Fig. 1): A bulk liquid region, consisting of the volatile liquid, a film region, where liquid films co-exist with saturated gas, and a dry region, consisting of only the unsaturated gas phase. By a combination of film flow and diffusion, the volatile liquid is transferred to the surface S, where it is purged. At the beginning of the process, the bulk liquid is in direct contact with the external gas, into which it evaporates directly.

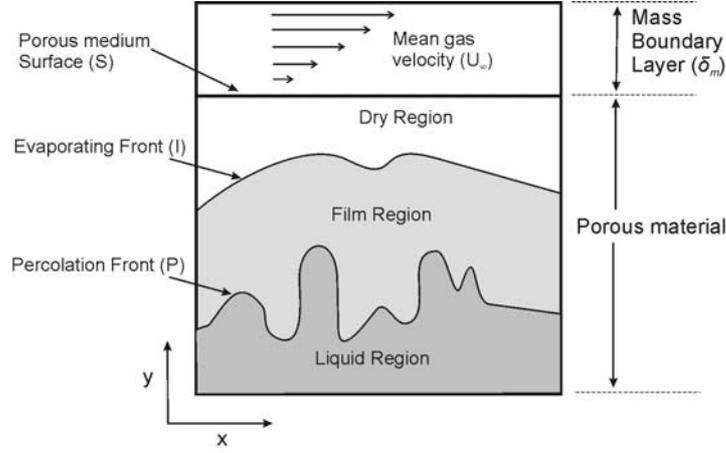


FIGURE 1. Schematic of the geometry considered for the drying of a porous medium. The volatile liquid evaporates within the porous medium and is transported by diffusion or through liquid films to the external surface S of the porous medium. A purge gas is flowing parallel to the surface and transfers the vapor by advection and diffusion over a mass transfer boundary layer of thickness δ_m .

The bulk liquid-gas interface (P) obeys Invasion Percolation rules, namely as the volatile liquid vaporizes, it recedes by vacating the largest perimeter throat it occupies at the given time. In this process, films are left in the corners of the pores, through which liquid can flow by capillarity to the evaporating front (I). The latter can be located either at the surface S or within the porous medium or partly at the surface S and partly within the porous medium, depending on whether films terminate at S or not. When the films terminate inside the porous medium, the liquid evaporates at the film tips and is transported towards S by gas-phase diffusion.

Inside the porous medium and under quasi-static conditions, the process in either the film region or in the unsaturated gas can be described by the Laplace equation (Yiotis et al., 2003, 2004). This is due to the fact that both for viscous flow in the films and for diffusion in the gas phase, the mass flux is linearly proportional to the driving force (pressure or concentration gradient, respectively). Then, the combined problem can be solved by defining the extended

variable $\Phi = \frac{\rho^3 + \zeta Ca_F}{1 + Ca_F}$, where ρ is the normalized thickness of the films (equal to one at the

percolation front), ζ is the gas-phase concentration of the volatile species, normalized with the equilibrium value (and thus equal to one at the percolation front) and Ca_F is a modified capillary number (see Yiotis et al., 2003 for the definition). Through this substitution, variable Φ can be shown to satisfy the Laplace equation in the entire domain, namely outside the percolation front and inside the porous medium (i.e. in the region bounded by surfaces P and S in Fig. 1)

$$\nabla^2 \Phi = 0 \quad (3)$$

When the product surface S is completely dry ($\zeta = 0$), corresponding to infinitely fast mass transfer, the problem reduces to solving (3) in the region outside the bulk liquid, but subject to boundary conditions $\Phi=1$ at the percolation front and $\Phi=0$ at S . From the solution obtained, one can determine in this case the position of the film tips, hence of the evaporation front I , by the condition $\Phi = \frac{Ca_F}{1 + Ca_F}$, which corresponds to $\rho=0$ and $\zeta=1$. The calculation of drying rates, film thickness profiles and other relevant quantities follow in a straightforward manner (Yiotis et al., 2004). However, in the general case, the external mass transfer is not infinitely fast, a dry interface cannot be maintained on the external boundary S , and the above approach does not apply. In that case the external mass transfer cannot become decoupled from the drying process inside the porous medium and a new approach is needed. This coupling is the main issue addressed in this paper.

2.1 Coupling of the external mass transfer with the porous medium

Consider the problem where the external mass transfer proceeds at finite rates, for example by advection along the x -direction and by diffusion along the x and y -directions (see Fig. 1). Inside the porous medium, the mechanisms discussed in Yiotis et al. (2003, 2004) remain valid, namely viscous flow in the films, diffusion in the gas phase, and the receding of the bulk liquid interface following percolation rules. Determining the film thickness and the concentration profiles will be solved by defining variable Φ as before, but by proceeding now in a different manner.

In the mass boundary layer outside the porous medium ($0 < y < d$) and under quasi-static conditions, the conservation of mass can be recast in the form

$$u \frac{\partial \Phi}{\partial x} = \frac{1}{Pe} \nabla^2 \Phi \quad \text{in} \quad 0 < x < n, \quad 0 < y < d \quad (4)$$

where we introduced the dimensionless velocity $u = \frac{U}{U_\infty}$, the two spatial coordinates were

scaled with a characteristic pore length scale l , and we defined the rescaled thickness $d = \frac{\delta_m}{l}$

and the pore-scale Peclet number, $Pe = \frac{U_\infty l}{D}$. Equation (4) is subject to a dry condition at the

edge of the boundary layer, $\Phi(x, y=d)=0$ and to periodic boundary conditions at $x=0$ and $x=n$ in the strictly diffusive case $Pe=0$. Inside the porous medium ($P(x, t) < y < 0$), the Laplace equation (3) remains applicable,

$$\nabla^2 \Phi = 0 \quad \text{in} \quad 0 < x < n, \quad P(x, t) < y < 0 \quad (5)$$

where we also defined the location of the percolation front, $y = P(x, t)$. Equations (4) and (5) are coupled as follows.

When bulk liquid interfaces or film menisci are absent on S , continuity on Φ and its normal derivative apply. When a bulk-liquid or a film interface exists at S , however, the solution is not as simple, as quantity Φ will be discontinuous at those points, and an iterative approach will be needed. Indeed, imposing the ‘‘naïve’’ film termination condition

$\Phi = \frac{Ca_F}{1 + Ca_F}$, for all films or liquid interfaces on S (a condition which is fully valid when the

films terminate inside the porous medium), renders the problem over-specified.

At the onset of drying, all pore throats at S are occupied by bulk liquid interfaces. During this stage, the drying rates are computed by solving the external mass transfer problem (4) using the saturated-gas boundary condition at S, namely $\Phi(x, y=0)=1$, and a dry-gas condition at the edge of the boundary layer, $\Phi(x, y=d)=0$. As drying proceeds, bulk-liquid interfaces will recede, creating liquid films at various surface pores. At any given time step, a surface pore containing a bulk-liquid interface may become a film-containing pore. Simultaneously, the film thickness ρ on other surface pores will decrease. Because of the possibility of discontinuities in Φ , finding the solution requires an iterative approach. The key is identifying the next surface pore where a bulk-liquid interface will recede and film will develop (and where Φ will have a jump), and also finding the respective jumps for all other surface pores containing films. This calculation is made using an iterative SOR algorithm, which ensures the following:

- If a surface pore does not contain films, condition $\Phi < \frac{Ca_F}{1 + Ca_F}$ applies and Φ is continuous across S at that point.
- If the surface pore contains films ($\Phi \geq \frac{Ca_F}{1 + Ca_F}$), then we set $\Phi = \frac{Ca_F}{1 + Ca_F}$ at this surface pore and look for the values of Φ of the neighbouring pores that satisfy the continuity of normal fluxes at the product surface S.

3. RESULTS AND DISCUSSION

We implemented the above approach to calculate the drying rates when the external mass transfer is coupled with the porous medium. The problem is characterized by the dimensionless parameters d (or δ_m), Pe , Ca_F , and n . In this paper we studied the effect of d (or δ_m), and Pe . The porous medium was represented as a cubic pore network with $n=50$ (see Yiotis et al., 2001 for details). This approach corresponds essentially to a finite-difference model of $50 \times 50 \times 50$ size.. As noted, lengths were made dimensionless with the lattice pore spacing, thus for example, $d=50$ means that the external boundary layer has the same extent as the pore network. First, we considered the solution of the problem in the absence of external flow where mass transfer is purely diffusive ($Pe=0$).

3.1 Effect of the diffusive mass transfer layer ($Pe=0$)

Figure 2 shows plots of the drying rate, normalized with respect to its maximum value (at the beginning of the process) vs. the overall liquid saturation for various values of the film thickness d and for a fixed value of $Ca_F=0.01$, in the limit when the external mass transfer is strictly diffusive ($Pe=0$). As the thickness of the external mass transfer layer increases, we find that a Constant Rate Period (CRP) emerges, the extent of which increases with d . For small values of d , e.g. for $d=2$, the films lose contact with the product surface soon after the beginning of the process and the product surface becomes dry. As a result, the drying rate

decreases relatively fast, as a function of the liquid saturation. At larger values, e.g. for $d=10$, however, the films remain in contact with the product surface for a sufficiently long time, until the liquid saturation drops below a critical value, corresponding to the Main Cluster Disconnection (MCD) point (shown in Fig. 2). Until that point, the drying rate remains constant. We note that in the above we have chosen a relatively high value for the capillary number Ca_F , as otherwise, for this specific lattice size, the films will remain long, spanning the entire pore network throughout the process, and the drying rates will remain constant for all values of the thickness. When d is small, the rate dependence is very similar to that reported by Le Bray and Prat (1999), and recently elaborated in Yiotis et al. (2006). In the latter, because of the absence of films, the existence of a constant-rate period requires that a significant fraction of the surface pores are occupied by bulk liquid-gas interfaces, so that the concentration at the surface remains close to its maximum (saturation) value. This mechanism is qualitatively different than when the boundary layer is thick, and where the CRP behavior emerges as a result of the existence of film-containing surface pores.

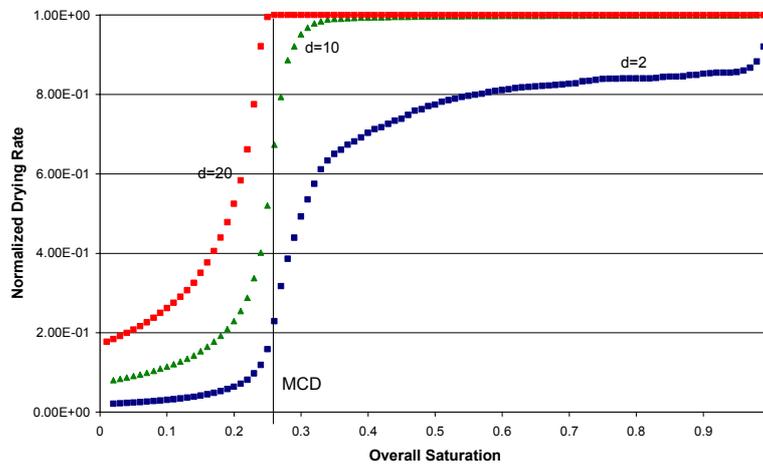


FIGURE 2. Normalized drying rate curves for various values of the boundary layer thickness d and for a fixed value of $Ca_F=0.01$ in the limit when the external mass transfer is strictly diffusive ($Pe=0$, $50 \times 50 \times 50$ pore networks). A Constant Rate Period (CRP) is clearly apparent for $d=10$ and $d=20$. For such relatively large value of Ca_F the films remain in contact with the product surface only when d is large. The curve corresponding to $d=2$ is similar to that reported by Le Bray and Prat (1999) in the absence of films.

In the absence of liquid films (Le Bray and Prat, 1999; Yiotis et al., 2006), namely when Ca_F takes large values, the extent of the CRP depends on the thickness d of the diffusive layer. This was first demonstrated by Suzuki and Maeda (1968) who solved the drying problem over a discontinuous source of liquid with respect to the length of the evaporating surface n and the thickness of the diffusive layer d . They showed that when the ratio $\varepsilon=n/d$ takes values less than 1, then the concentration contours vary almost linearly within the diffusive layer in the direction from the product surface to the side of the domain where the concentration is taken equal to zero and the drying rate remain practically constant, regardless of the saturation at the product surface (Fig. 3, right panel). When $\varepsilon>1$, then the drying rate

depends very strongly on the surface saturation (Fig. 3, left panel). Our results, shown in Fig. 2, agree qualitatively with the findings by Suzuki and Maeda (1968).



FIGURE 3. Concentration contours in the external layer over the surface of the porous medium corresponding to the Suzuki and Maeda (1968) problem for the strictly diffusive case ($Pe=0$) for two different values of the aspect ratio ε of the external layer. The liquid fraction at the product surface is 25%.

3.2 Effect of the Peclet number

The effect of the Peclet number was investigated by varying its value in the simulations, while keeping the thickness d and the capillary number Ca_F constant. For a number of different values of Pe tried, its direct effect was found to be negligible. This is due to the periodic boundary conditions applied along the flow x -direction, under the assumption that the unit cell considered is sufficiently far along the flow process. Because of the periodicity, $\frac{\partial\Phi}{\partial x}$ becomes zero for any value of Pe .

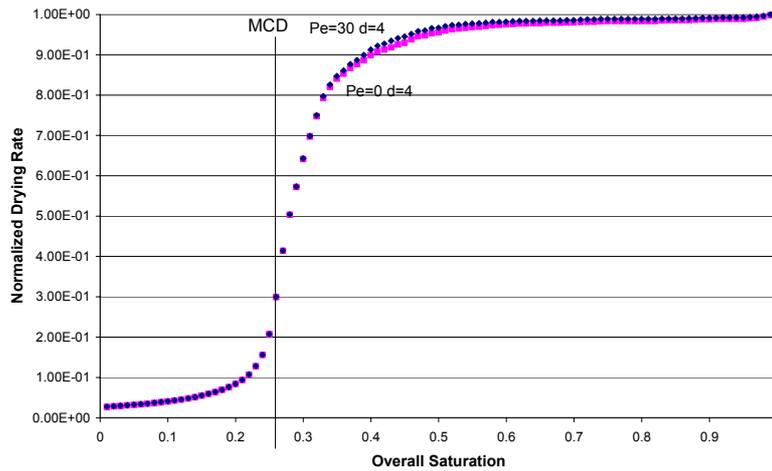


FIGURE 4. Normalized drying rate curves for different values of Pe when the velocity profile is piston-like. $Ca_F=0.01$, $d=4$.

Our results however were obtained under the simplifying assumption that the thickness of the mass boundary layer is independent of the Reynolds number. As shown in equation (2), higher values of Pe produce thinner mass boundary layers δ_m . This is expected to produce shorter CRPs with increasing Pe values (through thinner mass boundary layers), similar to the curves shown in Fig. 2.

The effect of the Reynolds number on the thickness of the mass boundary layer will be addressed in detail in a forthcoming publication.

4. CONCLUSIONS

In this paper, we present a coupled pore-network model that accounts for the mass transfer of the liquid vapors by diffusion and flow through liquid films within the porous medium, and convective mass transfer through the mass boundary layer over the surface of the porous medium. We study the effect of the thickness of the mass boundary layer and the Peclet number of the purge gas on the drying curves. Our results show that the drying rate remains practically constant as long as the liquid films span along the pore network and their density at the surface is sufficiently high. This condition is satisfied even for very low values of the residual liquid saturation when the thickness of the convective layer is greater than the average pore radius. The effects of the Peclet number and the velocity profile on the drying curves appear to be negligible.

REFERENCES

- Bird, R.B., W.E. Stewart and E.N. Lightfoot (1960), *Transport Phenomena*, John Wiley and Sons Ltd.
- Chen, P. and D.C.T. Pei, (1989), A mathematical model of drying processes, *Int. J. Heat Mass Transfer* 32(2), 297-310.
- Leal, G. (1992), *Laminar Flow and Convective Transport Process*, Butterworth-Heinemann.
- Le Bray, Y., Prat, M. (1999), Three-dimensional pore network simulation of drying in capillary porous media, *Int. J. Heat Mass Transfer* 42, 4207-4224.
- Suzuki, M. and S. Maeda, S. (1968), On the mechanism of drying of granular beds, *J. Chem. Eng. of Japan* 1(1), 26-31.
- Yiotis, A.G., A.K. Stubos, A.G. Boudouvis and Y.C. Yortsos (2001), A pore network model for drying processes in porous media, *Adv. Water Res.* 24(3), 439-460.
- Yiotis, A.G., A.G. Boudouvis, A.K. Stubos, I.N. Tsimpanogiannis and Y.C. Yortsos (2003), Effect of liquid films on the isothermal drying of porous media, *Phys. Rev. E* 68(3), 037303.
- Yiotis, A.G., A.G. Boudouvis, A.K. Stubos, I.N. Tsimpanogiannis and Y.C. Yortsos (2004), Effect of liquid films on the drying of porous media, *AIChE J.* 50(11), 2721-2737.
- Yiotis, A.G., A.K. Stubos, A.G. Boudouvis, I.N. Tsimpanogiannis and Y.C. Yortsos (2005), Pore network modelling of isothermal drying in porous media, *Transport Porous Media* 58(1-2), 63-86.
- Yiotis, A.G., I.N. Tsimpanogiannis, A.K. Stubos and Y.C. Yortsos (2006), Pore-network study of the characteristic periods in the drying of porous materials, *J Colloid Interface Science* doi:10.1016/j.jcis.2005.11.043.