

## Power law diffusion in drying processes

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**POWER LAW DIFFUSION  
IN  
DRYING PROCESSES**

W. J. COUMANS

**POWER LAW DIFFUSION  
IN  
DRYING PROCESSES**

# **POWER LAW DIFFUSION IN DRYING PROCESSES**

**PROEFSCHRIFT**

ter verkrijging van de graad van doctor  
aan de Technische Universiteit Eindhoven,  
op gezag van de rector magnificus,  
prof. dr. F. N. Hooge, voor een commissie  
aangewezen door het college van dekanen  
in het openbaar te verdedigen op  
vrijdag 29 mei 1987 te 14.00 uur

door

**WILHELMUS JOHANNES COUMANS**

geboren te Weert

Dit proefschrift is goedgekeurd door

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to my mother,

Marianne and Steven

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## CURRICULUM VITAE

The author was born on november, 15, 1945 at Weert, the Netherlands. After his secondary education and military service he joined Dutch State Mines (DSM) at Geleen in 1966. As a working student he followed a study for chemical analyst. After his examination in 1969 (cum laude) he started to work at Eindhoven University of Technology in the Polymer Engineering Group of Prof. D. Heikens. In 1970, again as a working student, he began his studies for chemical engineer at Eindhoven, which he concluded in 1978 (cum laude). His graduate research in the field of mechanical behaviour of plastic materials was performed in the group of Prof. D. Heikens. In 1980 he became the assistant of the late Prof. H.A.C. Thijssen, who stimulated him to start this research on drying in 1982.



## SUMMARY

In solids drying the calculation of drying rates and drying times is complicated by the concentration dependence of the diffusion coefficient and the shrinkage of the drying bodies due to the moisture loss. The calculation is further complicated if particles contain an internal gas core (e.g. in spray drying), which expands or shrinks during drying. In principle even the most complex processes can be calculated numerically. However, the lack of known product properties (for instance the relation between diffusion coefficient and moisture concentration) and the extremely difficult and laborious programming render the numerical approach prohibitive for practical application by process engineers and designers. It is the aim of this research to provide easily to handle *short-cut calculation procedures*, which are applicable to slabs, massive and hollow cylinders and spheres, irrespective of their degree of shrinkage. Diffusion of moisture in shrinking and non-shrinking bodies of the various geometries is described by a generalized diffusion equation. This equation has been solved numerically for a *power law dependence* of the diffusion coefficient on moisture concentration. Two boundary conditions, which are typical for two main drying stages, are considered: *constant surface flux* and *constant surface concentration*. Both drying stages can be subdivided into a *Penetration Period* (centre concentration hardly affected) and a *Regular Regime* (centre concentration clearly affected).

The numerical computer output can be described by simple correlations. For both Penetration Periods the introduction of a *G-parameter* appears to be helpful and for both Regular Regimes the concept of *Sherwood numbers* of the dispersed phase is of great importance. Drying times calculated with the correlations deviate less than 5% from the "exact" calculated values.

Moreover, a drying apparatus has been developed, to determine the weight of a sample (thin layer) as a function of time during drying (*drying curve*). The approximate correlations are used to evaluate the experimental drying curves for obtaining the relevant model parameters. Next the same correlations are used to predict drying curves at different drying conditions. Deviations between *predicted* and *experimental* drying curves of aqueous maltodextrin solutions appear to vary from a few percent to about 30% .

## SAMENVATTING

De berekening van het drooggedrag van deeltjes is erg gecompliceerd, omdat de diffusiecoëfficiënt van het migrerende vocht veelal sterk afhangt van het vochtgehalte. Een "exacte" berekening van het drooggedrag vereist kennis van numerieke methoden, grote vaardigheid in het programmeren en de beschikking over een main-frame computer. Bovendien is de relatie tussen de diffusiecoëfficiënt en het vochtgehalte meestal onbekend en zijn moeizame experimenten noodzakelijk ter bepaling hiervan. Proces ingenieurs en ontwerpers van droogapparatuur hebben in hun dagelijkse praktijk behoefte aan gemakkelijk te hanteren rekenprocedures.

Het onderzoek beschrijft een *kortsluitrekenmethode*, welke uitgaat van een machtsrelatie tussen de diffusiecoëfficiënt en het vochtgehalte (*power law diffusion*). De (cor)relaties van deze vereenvoudigde berekening zijn gebaseerd op de "exacte" numeriek berekende oplossingen. De afwijkingen in droogtijden, berekend volgens de "exacte methode" en de kortsluitmethode, bedragen gemiddeld 2% en zijn maximaal 5%. De kortsluitmethode is toepasbaar voor holle en massieve deeltjes met elke graad van krimp (vlakke lagen, cylinders en bollen).

Bovendien is een geautomatiseerde meetopstelling ontwikkeld, waarmee het gewicht van een monster (dunne laag) tijdens een droogproces kan worden geregistreerd (*droogcurven*). M.b.v. de vergelijkingen van de kortsluitmethode worden uit deze experimentele droogcurven de relevante model parameters berekend; vervolgens wordt met de kortsluitmethode het drooggedrag bij andere droogcondities voorspeld. De afwijkingen tussen voorspelde en experimentele droogtijden van maltodextrine oplossingen variëren van enkele procenten tot 30%.

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## CHAPTER I

### INTRODUCTION

Though drying is an economical important unit operation, which is frequently used in many branches of industry, it still is a hard to grasp process. The design of dryers is complicated by:

- the changing conveyance properties of the material during drying e.g. a wet droplet may first change into a sticky material and eventually into a free flowing powder;
- the prediction of the required drying time, which is a rather difficult calculation problem, especially if drying rates are fully controlled by diffusion of moisture in the material.

In this thesis the attention is focussed on the latter problem.

#### 1.1 Approximate Calculation Methods.

In solids drying mass transfer takes place from a dispersed phase (e.g. droplet of milk) to a continuous phase (e.g. air). Interface mass transfer processes are usually split up into mass transfer in the continuous phase and mass transfer in the dispersed phase.

*Mass transfer in the continuous phase* is generally described in a phenomenological way by means of a mass transfer coefficient (one film model). For some situations literature provides theoretical relations

for the calculation of mass transfer coefficients, however in most cases (semi-)empirical correlations of various dimensionless groups are to be used [1-9]. If mass transfer is fully controlled by the conditions of the continuous phase, the accuracy of the calculated drying rates is 20-30% at the best [3].

*Mass transfer in the dispersed phase depends on process conditions (e.g. mass transfer in the gas phase, temperature and humidity of the drying air) and product properties, such as:*

- *sorption-isotherm* [10,11], which describes the equilibrium between the thermodynamic moisture activity and the moisture content of the material;
- *parameters describing the various mass transfer mechanisms, such as molecular diffusion, capillary transport, etc.* [12,13]. In this thesis we confine ourselves to the description of rigid systems with a uniform temperature, thus internal circulations and temperature gradients do not contribute to mass transfer;
- *shrinkage behaviour* [14-16]; three cases can be distinguished:
  - maximum-shrinking*, if the decrease of the body volume equals the volume of the removed moisture;
  - partial-shrinking*, if the decrease of the body volume is less than the volume of the removed moisture;
  - non-shrinking*, if the volume of the body remains constant during drying.

In case of partial- and non-shrinking systems a porous solid will be formed during a drying process.

On the one hand, calculation models taking into account all mass transfer mechanisms with a maximum of physical relevance are generally not suited for practical use. On the other hand, easy to handle

calculation models with a too small physical basis often fail in predicting the drying behaviour with sufficient accuracy. Thus, in developing physical/mathematical models for the calculation of drying processes a good balance has to be found between the theoretical basis and the manageability of the calculation method.

Despite the occurrence of various mechanisms, quite often mass transfer is described with a single parameter, viz. the effective diffusion coefficient ( $D$ ), in which many mechanisms are lumped together. This rather rigid physical simplification implies that in general the diffusion coefficient will be strongly dependent on moisture content and temperature; a decrease of several orders of magnitude during drying may occur, e.g. for water in aqueous carbohydrate solutions [17-22]. For the mathematical description of the drying processes two main problems remain:

*first*, from a mass balance over an infinite small shell volume the diffusion equation is obtained. For constant diffusivity this equation has been solved analytically for many initial and boundary conditions. However, especially for particles with the geometry of a hollow cylinder or sphere the algebraic solutions are rather complicated and relatively difficult to handle (see Chapter IV). For concentration dependent diffusion coefficients a numerical approach is required. The numerical calculations are complicated, require highly sophisticated programming skill and high speed computers.

*second*, product properties, such as the relationship between diffusion coefficient and moisture content are rather inaccurate; deviations of 50% or more, depending on the range of moisture content, are quite common; moreover, in most cases product properties are even unknown and must be obtained from rather laborious experiments.

In general, *mathematically exact calculation procedures may be very complicated and require product properties, which however, are not exact, so there is a lot of room left for approximate calculation methods without substantial loss of overall accuracy.*

Approximate calculation procedures show the following advantages:

- first, they offer a better insight; simple equations will contribute to a better understanding of drying kinetics;*
- second, they are relatively easy to handle, especially of interest for practical engineers and designers;*
- third, the simple equations can be used in the evaluation of relevant product properties from laboratory drying experiments;*
- fourth, the approximate methods can also be useful in building up algorithms for controlling drying processes.*

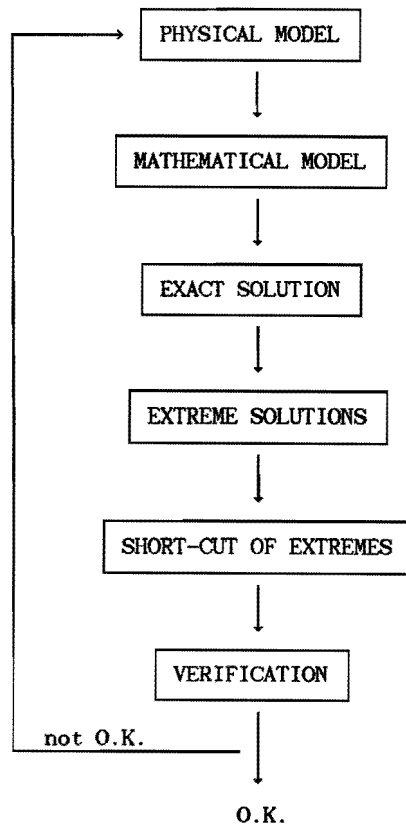
### 1.2 Strategy for Developing Approximate Calculation Methods.

In Figure 1.1 a strategy for developing approximate calculation methods is schematically depicted. First of all a description of the process in terms of physical phenomena is required. From this *physical model* a *mathematical model* can be derived. Usually the mathematical model consists of a partial differential equation with initial and boundary conditions. Next, we have to find the *exact solutions*. Sometimes these solutions can be found as algebraic expressions in literature, however in most cases one has to resort to a numerical approach. For *extreme situations* (e.g. short times, large times, low flux, high flux) quite often the exact solutions reduce to a simple form. Then the approximate solutions of the two extremes are connected by means of a simple correlation (*short-cut of extremes*, therefore the approximate methods are often referred to as *short-cut*



calculation methods). Finally, the approximate solutions are compared with the exact solutions. The result of this *verification* determines whether one has to make corrections or not.

It will be shown in this thesis that by tolerating small errors ( $\leq 5\%$ ) tremendous simplifications of the calculation procedure can be achieved.



**Figure 1.1** Strategy for developing short-cut calculation methods.

### 1.3 Short History

This research on approximate calculation methods for drying processes is a continuation of the investigations carried out by Schoeber [23,24], Luyben, Olieman, Bruin and Liou [25-30]. Similar research on drying kinetics was later started by Yamamoto, Sano and Hoshika [31,32].

Schoeber analyzed the numerical solutions of the diffusion equation for many kinds of concentration dependence of the diffusion coefficient. From the numerical solutions he developed general approximate methods for the calculation of mass transfer in diffusion processes. Schoeber's concept is based on the combination of "short time solutions" (Penetration Period) and "large time solutions" (Regular Regime). It appears from his analysis that the Regular Regime solutions can serve as a basis for the evaluation of diffusion coefficients from experimental drying curves.

Luyben measured the drying curves of a number of food materials and compared them with calculated drying curves obtained from short-cut calculations (based on Schoeber's approach) and "exact" numerical calculations. Furthermore, based on Schoeber's equations, he developed a method to derive diffusion coefficients from non-isothermal drying curves of bodies with various geometries (slabs, cylinders and spheres).

The concentration dependence of the diffusion coefficient can be described in many ways: exponential, logarithmic, power law, polynomial, etc.. However, none of them will really describe in a satisfying way the experimental values of the diffusion coefficient of various systems over the whole concentration range of interest, unless one resorts to equations with many (more than two) fitting parameters.

Based on the work of Schoeber and Luyben a power law dependence of the effective diffusion coefficient ( $D$ ) with concentration was put forward

by Liou and Bruin:

$$D\rho_s^2 = b \left[ \frac{\rho_m}{\rho_s} \right]^a \quad (1.1)$$

in which,  $D$  is diffusion coefficient [ $m^2/s$ ],  $\rho_m$  and  $\rho_s$  are the concentrations of moisture and solid [ $kg/m^3$ ],  $a$  and  $b$  are fitting parameters.

Liou proposed a short-cut calculation method for drying rates, drying times and concentration profiles. His method is valid for non-shrinking and maximum-shrinking slabs, massive cylinders and massive spheres with a constant surface concentration.

Liou's approximate method for "power law diffusion" has been extended to hollow and massive systems with any degree of shrinkage and to systems with constant surface flux by Coumans and Thijssen [33,34]; this approach will be described systematically in this thesis.

#### 1.4 Drying Stages.

The mathematical formulation of the external boundary condition of the diffusion equation depends on the drying stage. In this thesis three main stages are distinguished (see also Chapter IV):

Stage I, at the beginning of the drying process the surface moisture activity remains nearly constant. For slabs and non-shrinking cylinders and spheres this period is characterized by a nearly constant surface flux.

Stage II, both the boundary flux and the boundary concentration decrease considerably during this stage. The boundary condition now is more complex and requires good knowledge of the sorption-isotherm.

Stage III: the surface concentration, relative to its total change, hardly changes anymore. Thus, by good approximation, there is a constant surface concentration.

All three main stages can be subdivided into two substages:

Penetration Period (for short times), which is characterised by a centre concentration which has hardly changed.

Regular Regime (for large times), during which the centre concentration, compared to its initial value, significantly decreases; the most striking feature of this drying stage is that *the drying behaviour has become independent of the initial conditions.*

It appears that every substage has its own approximate equations.

This thesis only deals with drying stages I and III. It is assumed that drying stage II, which is rather hard to describe, has a minor contribution to the total drying time (see Chapter VI); therefore drying stage II has not been investigated. Approximate methods for the calculation of the surface concentration as function of time (stage I) and for the calculation of drying rates and drying times at any given value of the averaged moisture concentration (stage III) have been derived from the analytical and numerical solutions of the diffusion equation.

The short-cut equations of drying stage I will be of special interest e.g. for spray drying processes [21] in order to predict whether droplets will agglomerate or not, or whether they will stick to the wall of the dryer or not. Also the application of the method in short-cut calculation methods for the prediction of aroma losses during drying [35,36] is interesting. The equations of drying stage III will be advantageous in predicting overall drying times, since for high intensity drying this stage contributes dominantly to the total

drying time.

The described short-cut methods refer to mass transfer in dispersed phases, where diffusion plays an important role and even may become rate controlling; though this thesis emphasizes drying processes the methods described may also be applied to processes such as leaching, absorption, humidification and desorption.

### 1.5 Scope of this Thesis.

The contents of the chapters are briefly summarized below.

Chapter II: Based on a comparison of mass transfer in maximum- and non-shrinking systems, a generalized mass transfer equation is formulated for systems with any degree of shrinkage. Further some aspects of mass and heat transfer coefficients in the gas phase are considered.

Chapter III: Mass transfer in slabs, hollow and massive cylinders and spheres with any degree of shrinkage is described with a *generalized diffusion equation*. By transforming this equation into a dimensionless form, a similarity of solutions is obtained. Also a generalized representation of the total mass balance is presented.

Chapter IV: Approximate equations are derived from the analytical solutions of *non-shrinking systems with constant diffusivity*. It appears that this most simple drying situation yields approximate equations, which only need slight adaptations for shrinking systems with variable diffusion coefficients.

Chapter V: For power law diffusion in systems with any degree of shrinkage approximate equations are derived from the numerical solutions.

Chapter VI: An experimental set-up for the continuous registration of the weight of a drying slab as function of time is described. Several drying experiments have been performed with thin layers of aqueous maltodextrin solutions. From the so-called *drying curves* (=sample weight versus time) relevant physical information is derived by means of the approximate equations of Chapter V. Finally, drying curves are predicted and compared with the experimental ones.

## CHAPTER II

### MASS AND HEAT TRANSFER IN DRYING PROCESSES

#### 2.1 Introduction.

Mass transfer in drying bodies is often described by means of an effective diffusion coefficient. In literature one comes across many definitions of the diffusion coefficient and the mass transfer equations can take on many forms. Sometimes the definitions used are somewhat obscure.

The aim of this chapter is to define in an unambiguous way the basic equations for mass transfer in well-defined binary systems.

Unfortunately, in most cases drying systems are thermodynamically ill-defined. Further, extra complications are caused by shrinkage due to the loss of moisture during drying. Therefore a comparison between shrinking and non-shrinking systems is given, leading to a generalized definition of the mass transfer equation for systems irrespective of their shrinkage behaviour.

The drying process is induced by the conditions of the external gas phase, especially the temperature, the humidity and the mass- and heat-transfer coefficients. The last part of this chapter deals with some aspects of these mass- and heat-transfer coefficients relevant for drying processes.

## 2.2 Mass Transfer in Ideal One-Phase Systems.

In one-phase systems, like gas mixtures and liquid solutions, two mechanisms for mass transfer can be distinguished:

- molecular diffusion caused by a concentration gradient;
- convection caused by a pressure gradient (forced convection) or a vertical density gradient (free convection); bulk flow may also be induced by a concentration gradient.

It is important to realize that the contribution of each mechanism depends on the definition of the coordinate system with respect to which mass fluxes are defined. In a coordinate system, fixed with respect to the observer, the mass flux  $n_i$  of a component  $i$  with concentration  $\rho_i$  and velocity  $v_i$  is given by:

$$n_i = \rho_i v_i \quad (2.1)$$

The mass flux  $n_i$  is built up of a convective flux and a diffusion flux  $J_i$ , which is defined with respect to a certain reference velocity  $v_{ref}$ . Equation 2.1 can also be written as:

$$n_i = \rho_i v_{ref} + \rho_i (v_i - v_{ref}) \quad (2.2)$$

or

$$n_i = \rho_i v_{ref} + J_i \quad (2.3)$$

The reference velocity  $v_{ref}$  can be defined as:

- 1) the mass average velocity  $v$ :

$$v = \frac{\sum_i \rho_i v_i}{\sum_i \rho_i} = \frac{\sum_i n_i}{\rho} \quad (2.4)$$



where  $\rho$  is the total mass concentration of the mixture.

ii) the volume average velocity  $v^{\square}$ :

$$v^{\square} = \frac{\sum_i \frac{\rho_i}{d_i} v_i}{\sum_i \frac{\rho_i}{d_i}} = \frac{\sum_i n_i}{\sum_i d_i} \quad (2.5)$$

where  $d_i$  is the partial density of component  $i$ ,

$\rho_i/d_i$  is the volume fraction of component  $i$  in the mixture and

$n_i/d_i$  is the volume flux of  $i$ .

iii) the velocity of another component in the mixture, for instance of

component  $b$ , thus  $v_{\text{ref}} = v_b$ .

iv) molar average velocity  $v^{\circ}$ :

$$v^{\circ} = \frac{\sum_i c_i v_i}{\sum_i c_i} = \frac{\sum_i N_i}{c} \quad (2.6)$$

where  $c_i$  is the molar concentration of component  $i$ ,

$c$  is the total molar concentration of the mixture and

$N_i$  is the molar flux of component  $i$ .

Most frequently the mass average velocity is chosen as a reference for the diffusion flux. Thus for a binary mixture with components  $a$  and  $b$  we get for the convective flux of  $a$ :

$$\rho_a v = \rho_a \frac{n_a + n_b}{\rho} = (n_a + n_b) \omega_a \quad (2.7)$$

where  $\omega_a$  is the mass fraction of component  $a$  on total basis.

The diffusion flux  $j_a$  is described according to Fick's law with a

diffusion coefficient  $D_a$  :

$$j_a = -D_a \rho \frac{\partial \omega_a}{\partial r} \quad (2.8)$$

From equations 2.3, 2.7 and 2.8 the basic equation for mass transfer in binary solutions follows:

$$n_a = (n_a + n_b) \omega_a - D_a \rho \frac{\partial \omega_a}{\partial r} \quad (2.9)$$

In fact equation 2.9 defines the diffusion coefficient  $D_a$  of component a in a binary mixture. It can simply be proved that  $D_a = D_b = D$ .

Taking another reference velocity will change the separate contributions of convection and molecular diffusion to the total mass flux. In Table 2.1 a survey is given of expressions for the mass flux. It should be realised that in all these equations the diffusion coefficient  $D$  is defined according to equation 2.9. Further, all these equations are equivalent, because they all describe the same mass flux. However, in some special cases a sensible choice will lead to a simplification of the flux expression, namely:

- |  |   |
|--|---|
| i) no net mass flux ( <i>equimass</i> )          | : $n_a + n_b = 0 \longrightarrow$ choose I                          |
| ii) no net volume flux ( <i>equivolumetric</i> ) | : $\frac{n_a}{d_a} + \frac{n_b}{d_b} = 0 \longrightarrow$ choose II |
| iii) Stefan diffusion                            | : $n_b = 0 \longrightarrow$ choose I, II<br>III or IV               |
| iv) no net molar flux ( <i>equimolar</i> )       | : $N_a + N_b = 0 \longrightarrow$ choose IV                         |

Strictly, the equations of Table 2.1 only apply to binary one-phase systems, however most drying systems are multi-component systems and consist of more than two phases.

**Table 2.1** Survey of expressions for the massflux of component a in a binary mixture.

reference velocity	total mass flux = convective flux + diffusive flux
I mass average	$n_a = (n_a + n_b)\omega_a - D\rho \frac{\partial \omega_a}{\partial r}$
II volume average	$n_a = \left[ \frac{n_a}{d_a} + \frac{n_b}{d_b} \right] \rho_a - D \frac{\partial \rho_a}{\partial r}$
III velocity of b	$n_a = n_b \frac{\omega_a}{1-\omega_a} - \frac{D\rho}{1-\omega_a} \frac{\partial \omega_a}{\partial r}$
IV molar average	$N_a = (N_a + N_b)x_a - Dc \frac{\partial x_a}{\partial r}$
<p><u>Help equations:</u></p> <p>1. <math>\rho_a + \rho_b = \rho</math>      2. <math>\omega_a = \frac{\rho_a}{\rho}</math>      3. <math>\omega_a + \omega_b = 1</math></p> <p>4. <math>c_a + c_b = c</math>      5. <math>x_a = \frac{c_a}{c}</math>      6. <math>x_a + x_b = 1</math></p> <p>7. <math>\omega_a = \frac{\rho_a}{\rho} = \frac{\rho_a}{\rho_a + \rho_b} = \frac{\rho_a / \rho_b}{1 + \rho_a / \rho_b}</math></p>	

### 2.3 Mass Transfer in Real Systems.

In drying processes three types of systems can be distinguished: maximum-shrinking, partial-shrinking and non-shrinking systems.

Henceforth it is assumed that shrinkage, if present, is isotropic (e.g. the shape of a shrinking body does not change) and that partial densities are constant.

### 2.3.1 Maximum-Shrinking Systems.

Shrinking is maximum if the volume decrease of the drying material equals the volume of the removed moisture (Examples: sugar solutions, milk, coffee extract, fruit juices). It is true that these systems are seldom binary, however, with respect to the drying process they may be considered as such with components moisture(m) and dissolved solids(s). In these systems the volume balance reads:

$$\frac{\rho_m}{d_m} + \frac{\rho_s}{d_s} = 1 \quad (2.10)$$

where  $\rho_m/d_m$  and  $\rho_s/d_s$  are the volume fractions of moisture and dissolved solids respectively. Mass transfer of moisture and solids occurs by means of convection and molecular diffusion, so the equations of Table 2.1 may be applied. The basic mass transfer equation now reads:

$$n_m = (n_m + n_s)\omega_m - D\rho \frac{\partial \omega_m}{\partial r} \quad (2.11)$$

Because no pores are formed during drying, there is no net volume flux in maximum shrinking systems, thus also:

$$n_m = -D \frac{\partial \rho_m}{\partial r} \quad (2.12)$$

An expression for the moisture diffusion flux  $j_m^s$  with respect to the solids velocity  $v_s$  is found as follows:

$$n_m = \rho_m v_s + j_m^s \quad (2.13)$$

Also

$$n_s = \rho_s v_s \quad (2.14)$$

Eliminating  $n_m$  and  $n_s$  from equations 2.11, 2.13 and 2.14 gives:

$$j_m^s = - \frac{D\rho}{1-\omega_m} \frac{\partial \omega_m}{\partial r} \quad (2.15)$$

With the help equations of Table 2.1 this expression can be transformed into:

$$j_m^s = - D\rho_s \frac{\partial(\rho_m/\rho_s)}{\partial r} \quad (2.16)$$

It should be noticed, that  $\rho_m/\rho_s$  is the so-called solids based moisture concentration (kg m/kg s).

### 2.3.2 Non-Shrinking Systems.

Non-shrinking systems are characterized by a constant volume during drying. This means that the volume of the moisture, which has been removed from the system, will be replaced by an equal volume of gas. (Examples: air-drying of ceramics and sand.)

During the drying process of such systems a capillary porous material is built up, consisting of:

- three components viz. moisture (m), solid (s) and air (a),
- several phases, viz. a solid matrix (S) with pores containing liquid moisture (L) and/or moisture vapour (G) and/or air (G).

Mass transfer of moisture in capillary porous materials may occur by several mechanisms [12,13], such as capillary liquid flow, molecular diffusion, evaporation-condensation, surface diffusion, etc.. During a particular stage of the drying process one of these mechanisms may

dominate, but in general several mechanisms will be present simultaneously. A fundamental description of mass transfer based on all occurring mechanisms will lead to rather complicated physical/mathematical models.

In a rigorous but more practical approach the non-shrinking material is considered as a pseudo one-phase system with three components viz. moisture(m), solid(s) and air(a). The volume fraction of the gaseous components (air/vapour) is called the porosity  $\epsilon$ . Assuming that a negligible part of the moisture is present as vapour the volume balance reads:

$$\frac{\rho_m}{d_m} + \frac{\rho_s}{d_s} = 1 - \epsilon \quad (2.17)$$

Similar to a binary system the diffusion coefficient of this ternary system is defined as:

$$n_m = (n_m + n_s + n_a) \omega_m - D\rho \frac{\partial \omega_m}{\partial r} \quad (2.18)$$

Because a certain volume of liquid (moisture) is replaced by an equal volume of gas (air) the averaged moisture flux will be about 1400 times larger than the averaged air flux, therefore:

$$n_m \approx (n_m + n_s) \omega_m - D\rho \frac{\partial \omega_m}{\partial r} \quad (2.19)$$

In non-shrinking systems a rigid non-moving solid matrix exists ( $n_s=0$ ), so that:

$$n_m = - \frac{D\rho}{1 - \omega_m} \frac{\partial \omega_m}{\partial r} \quad (2.20)$$

and from equation 2.13:

$$n_m = j_m^s \quad (2.21)$$

Taking into account that the solid concentration is constant ( $\rho_s = \rho_{s0}$ ) and that

$$\omega_m = \frac{\rho_m}{\rho} = \frac{\rho_m}{\rho_m + \rho_s + \rho_a} \approx \frac{\rho_m}{\rho_m + \rho_s} \quad (2.22)$$

equation 2.20 can be transformed into:

$$n_m = j_m^s = -D \frac{\partial \rho_m}{\partial r} = -D \rho_s \frac{\partial (\rho_m / \rho_s)}{\partial r} \quad (2.23)$$

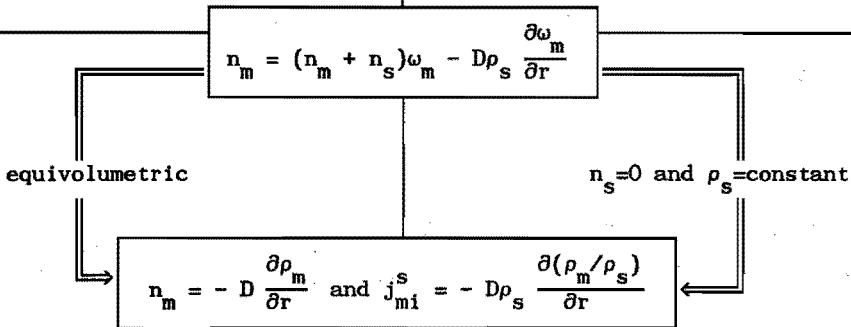
From the above it can be stated that a non-shrinking system can also be looked upon as a pseudo binary system (m+s). However, it is not meaningful to speak now in terms of equivolumetric mass transfer or not, because mass transfer of moisture may occur both as vapour and liquid. Further, the parameter D should be considered now as an effective diffusion coefficient, in which the contributions of several mass transfer mechanisms are lumped together.

### 2.3.3 Systems with Any Degree of Shrinkage.

Characteristics of maximum-shrinking and non-shrinking systems are summarized once more in Table 2.2. From this table it can be seen that the same mass transfer equations apply for two extremes in shrinking behaviour, so it is obvious to apply these equations also for intermediate shrinking behaviour. Thus for all systems, regardless

**Table 2.2** Comparison of mass transfer in maximum-shrinking and non-shrinking systems.

MAXIMUM-SHRINKING SYSTEM	NON-SHRINKING SYSTEM
mass transfer in one phase	mass transfer in more phases
pseudo binary system (m+s)	pseudo ternary (m+s+gas) or pseudo binary (m+s) system
migrating components in solution	migrating components may occur in several aggregate states
two mechanisms of mass transfer	several mechanisms of mass transfer
volume balance: $\frac{\rho_m}{d_m} + \frac{\rho_s}{d_s} = 1$	volume balance: $\frac{\rho_m}{d_m} + \frac{\rho_s}{d_s} = 1 - \epsilon$
equivolumetric mass transfer: $\frac{n_m}{d_m} + \frac{n_s}{d_s} = 0$	non-moving solid matrix: $n_s = 0 \text{ and } \rho_s = \rho_{s0} = \text{constant}$
practical and thermodynamically meaningful definition of the diffusion coefficient:	practical but thermodynamically meaningless definition of the diffusion coefficient:





their degree of shrinkage, equation I (see Table 2.1) is taken as the generalized basic equation for mass transfer:

$$n_m = (n_m + n_s) \omega_m - D \rho \frac{\partial \omega_m}{\partial r} \quad (2.24)$$

Because equations 2.13 and 2.14 are general, it follows that equation 2.15 gives a general relation for the diffusion flux with respect to the solids velocity; applying equation 2.22 then yields:

$$j_m^s = - D \rho_s \frac{\partial (\rho_m / \rho_s)}{\partial r} \quad (2.25)$$

The generalized volume balance reads:

$$\frac{\rho_m}{d_m} + \frac{\rho_s}{d_s} = 1 - \epsilon \quad (2.26)$$

where the porosity  $\epsilon$  not only depends on moisture concentration but also on the degree of shrinkage. The relationship among porosity, moisture concentration and degree of shrinkage will be worked out in Chapter III.

Irrespective of the shrinkage behaviour, it appears that the basic mass transfer equation 2.24 can also be transformed into:

$$n_m = -D \frac{\partial \rho_m}{\partial r} \quad (2.27)$$

This result seems to be trivial, however it is not, because it can not be seen beforehand that equations 2.24, 2.25 and 2.27 are also consistent for systems irrespective their shrinkage behaviour.

## 2.4 Simultaneous Heat and Mass Transfer.

For a description of the external mass transfer of a drying body one has to deal correctly with the simultaneously occurring heat transfer from the gas phase towards the body. A sophisticated approach for this problem can be found in literature [8,9]. However, in this section simple basic equations for mass and heat transfer in the gas phase will be given.

### 2.4.1 Heat Flux and Heat Transfer Coefficient.

In most convective dryers heat transfer takes place to bodies submerged in a relatively extended gas phase, mostly air. It is assumed that the resistance for heat transfer is concentrated in a thin film at the interface  $i$  (Figure 2.1). In this film, with thickness  $\delta_H$ , the temperature of the gas phase increases from  $\theta'_i$  at the interface to  $\theta'_\infty$  in the bulk.

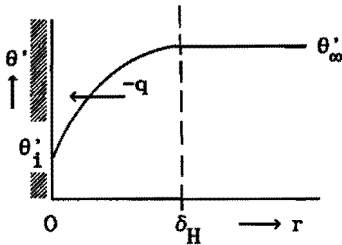


Figure 2.1 Schematic representation of film for heat transfer.

Consider now steady-state conditions for mass and heat transfer. The heat flux  $q$  in the film consists of a convective heat flux caused by the moisture flux and a conductive heat flux due to the temperature gradient:

$$q = n_m c_{pv} (\theta' - \theta'_i) - \lambda \frac{\partial \theta'}{\partial r} \quad (2.28)$$

where  $c_{pv}$  is the heat capacity of the vapour; the diffusive contribution is written by analogy with the Fourier law of heat conduction, however, the quantity  $\lambda'$  is now the so called "turbulent coefficient of thermal conductivity" or "Eddy Conductivity" [1]. It is important to realize that  $\lambda'$  is not a physical property of the gas phase, but depends on position, direction and the nature of the turbulent flow in the film.

Because of steady-state conditions equation 2.28 can now be integrated between the limits  $r=0$  (interface) and  $r=\delta_H$  (bulk). In case of no mass flux ( $n_m=0$ ) integration yields:

$$q = \frac{1}{\delta_H \int_0^{\delta_H} \frac{\partial r}{\lambda'}} (\theta'_i - \theta'_\infty) \quad (2.29)$$

The heat transfer coefficient  $\alpha'$  is defined according to:

$$\frac{1}{\alpha'} = \int_0^{\delta_H} \frac{\partial r}{\lambda'} \quad (2.30)$$

and thus equation 2.29 now becomes:

$$q = \alpha' (\theta'_i - \theta'_\infty) \quad (2.31)$$

For an adiabatic drying process ( $n_m \neq 0$ ) it is assumed that all the heat reaching the interface is used for evaporation of the moisture, thus:

$$q + n_m L_i = 0 \quad (2.32)$$

where  $L_i$  is the enthalpy of evaporation at  $\theta'_i$ .

Elimination of  $n_m$  from equations 2.28 and 2.32 and next integration

yields:

$$q = -\alpha' \frac{L_i}{c_{pv,f}} \ln \left[ 1 + \frac{c_{pv,f}}{L_i} (\theta'_\infty - \theta'_i) \right] \quad (2.33)$$

where  $c_{pv,f}$  is the heat capacity of the vapour at average film conditions.

The effective heat transfer coefficient ( $\alpha'_{eff}$ ) is defined as:

$$q = \alpha'_{eff} (\theta'_i - \theta'_\infty) \quad (2.34)$$

Thus:

$$\frac{\alpha'_{eff}}{\alpha'} = \frac{\ln \left[ 1 + \frac{c_{pv,f}}{L_i} (\theta'_\infty - \theta'_i) \right]}{\frac{c_{pv,f}}{L_i} (\theta'_\infty - \theta'_i)} \quad (2.35)$$

In many practical drying situations  $\frac{c_{pv,f}}{L_i} (\theta'_\infty - \theta'_i) \ll 1$  and a first approximation of equation 2.35 is given by:

$$\frac{\alpha'_{eff}}{\alpha'} = 1 - \frac{1}{2} \frac{c_{pv,f}}{L_i} (\theta'_\infty - \theta'_i) \quad (2.36)$$

Quite often it even holds that  $\alpha'_{eff} \approx \alpha'$ . It can be concluded now that in many practical drying situations the influence of mass transfer on the heat transfer coefficient  $\alpha'$  will be negligible.

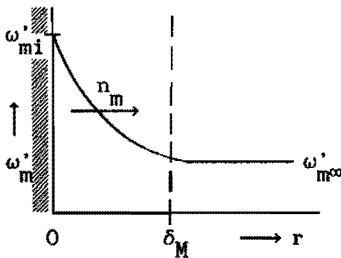
Values of  $\alpha'$  are calculated from (semi-) empirical correlations for heat transfer [1-9], which take in general the following form:

$$Nu = c_1 + c_2 Re^n Pr^m \quad (2.37)$$

where  $Nu$  is Nusselt number,  $Re$  is Reynolds number,  $Pr$  is Prandtl number,  $c_1, c_2, n, m$  are (semi-)empirical constants. In Table 2.3 the definitions of  $Nu$ ,  $Re$  and  $Pr$  are summarized.

#### 2.4.2 Mass Flux and Mass Transfer Coefficient.

Consider steady-state mass transfer from the surface of a drying material to an unsaturated drying gas. It is generally assumed, that the resistance for mass transfer is concentrated in a thin film close to the interface (Figure 2.2).



**Figure 2.2** Schematic representation of film for mass transfer.

The gas phase may be seen as a pseudo binary one-phase system with components moisture(m) and air(a).

In a way similar to the heat transfer coefficient (§ 2.4.1), the mass transfer coefficient  $k'$  in the gas phase is defined by:

$$\frac{1}{k'} = \int_0^{\delta_M} \frac{dr}{D'} \quad (2.38)$$

With shrinking systems no air is transferred across the interface.

With non-shrinking or partial-shrinking systems the air flux through the interface will be negligible with respect to the moisture flux (§ 2.3.2). So, the mass transfer of moisture in the film can be described as a Stefan diffusion process:

$$n_m = \frac{D' \rho'}{1 - \omega'_m} \frac{\partial \omega'_m}{\partial r} \quad (2.39)$$

By integrating this equation a problem arises. In most practical cases the moisture concentration will be very low, so that  $\rho'$  is hardly affected by the concentration. However, because of simultaneous heat transfer a temperature gradient in the film exists and  $\rho'$  will be a function of space  $r$ . Finding the space dependence of  $\rho'$  is a rather complicated problem. One can get around this difficulty by putting a constant value  $\rho'_f$ , which is defined as the total concentration at the average film conditions  $\theta'_f$  and  $\omega'_f$ , where:

$$\theta'_f = \frac{\theta'_i + \theta'_\infty}{2} \quad \text{and} \quad \omega'_f = \frac{\omega'_i + \omega'_\infty}{2} \quad (2.40)$$

Integration of equation 2.39 now yields the expression for the mass flux in the film:

$$n_m = k' \rho'_f \ln \left[ \frac{1 - \omega'_{m,\infty}}{1 - \omega'_{m,i}} \right] \quad (2.41)$$

The effective mass transfer coefficient  $k'_{\text{eff}}$  is defined as:

$$n_m = k'_{\text{eff}} \rho'_f (\omega'_{mi} - \omega'_{m\infty}) \quad (2.42)$$

From equations 2.41 and 2.42 follows:

$$\frac{k'_{\text{eff}}}{k'} = \frac{\ln \left[ \frac{1 - \omega'_{m\infty}}{1 - \omega'_{mi}} \right]}{\omega'_{mi} - \omega'_{m\infty}} \quad (2.43)$$

At not too high temperatures of the interface (e.g.  $\theta_i' \leq 50^\circ\text{C}$ ) the moisture concentrations in the film will be very low ( $\omega_{mi}' \ll 1$  and surely  $\omega_{m\infty}' \ll 1$ ) and a first approximation of equation 2.43 is:

$$\frac{k'_{\text{eff}}}{k'} \approx \frac{1}{1 - \omega_{mi}'} \quad (2.44)$$

and in many practical drying situations even:  $k'_{\text{eff}} \approx k'$ .

The mass transfer coefficient  $k'$  is found from (semi-)empirical correlations, which can be obtained by analogy with the heat transfer correlations (Table 2.2). In general, the correlations for steady-state mass transfer to bodies, submerged in an extensive fluidum take on the following form (see also equation 2.37):

$$\text{Sh} = c_1 + c_2 \text{Re}^n \text{Sc}^m \quad (2.45)$$

where Sh is the Sherwood number for the gas phase and Sc is the Schmidt number.

**Table 2.3** Analogy between Heat and Mass Transfer.

heat transfer			mass transfer		
number	symbol	definition	number	symbol	definition
Nusselt	Nu	$\frac{\alpha' l}{\lambda'}$	Sherwood	Sh	$\frac{k' l}{D'}$
Reynolds	Re	$\frac{v' l}{\nu'}$	Reynolds	Re	$\frac{v' l}{\nu'}$
Prandtl	Pr	$\frac{\nu'}{a'}$	Schmidt	Sc	$\frac{\nu'}{D'}$





## CHAPTER III

### GENERALIZED FORMULATION OF DIFFUSION EQUATION AND MASS BALANCE

#### 3.1 Physical Model.

The physical model used in this thesis is based on the concept of a concentration dependent diffusion coefficient, which accounts for the fact that not all of the moisture is bound equally strong or through the same physical mechanisms. Considered is the drying of shrinking and non-shrinking bodies with the following standard geometries: the infinite slab, the infinite (hollow) cylinder and the (hollow) sphere.

Postulates underlying the physical model are as follows:

1. The particle has a uniform temperature, which does not change with time (isothermal drying).
2. The particle consists of two or three components:
  - solid component (e.g. milk solids);
  - moisture component (e.g. water);
  - gas component (e.g. air), replacing the removed moisture in case of not completely shrinking systems.
3. Mass transfer in the particle, however, is regarded as a pseudo binary diffusion process (moisture and solid), for which, irrespective of the degree of shrinkage, the mass transfer

equations of §2.2.3 may be applied.

4. The moisture diffusion coefficient depends on the moisture content; for practical calculations a power-law relationship is proposed. The temperature dependence of the diffusion coefficient can be described by means of an Arrhenius type relation.
5. The volume reduction of shrinking systems is:
  - linear proportional to the moisture loss;
  - isotropic (no deformation of the body);
  - homogeneous (the local porosity in the moisture free solid is independent of the space coordinates).
6. The partial quantities (e.g. densities) of the constituting components are independent of the changing composition.
7. The moisture flux towards the gas phase, enclosed in hollow bodies, is assumed to be zero.
8. The size of the enclosure, in case of hollow bodies, remains constant.
9. Diffusion can be described by using only one space coordinate.
10. Drying starts with a body of homogeneous composition, not considering the hollow core if any.
11. The external boundary condition is determined by the equilibrium sorption properties of the material being dried and the conditions of the gas medium carrying off the moisture. Two special cases will be considered:
  - constant surface water activity (e.g. in case of non-shrinking bodies this means a constant surface flux);
  - constant surface water concentration (e.g. surface of the drying body close to equilibrium with the external gas medium).

### 3.2 Mathematical Model.

The diffusion equation describing moisture transfer follows from a mass balance over an infinite small shell volume:

Partial differential equation:

$$\frac{\partial \rho_m}{\partial t} = \frac{1}{r^v} \frac{\partial}{\partial r} (r^v n_m) \quad (3.1)$$

Initial condition:

$$t=0 \quad R_1 < r < R_2(0) \quad \rho_m = \rho_{m0} \quad (3.2)$$

Boundary conditions:

$$t > 0 \quad r = R_1 \quad \frac{\partial(\rho_m/\rho_s)}{\partial r} = 0 \quad (3.3)$$

$$r = R_2(t) \quad \rho_m = \rho_{mi}(t) \quad (3.4)$$

or

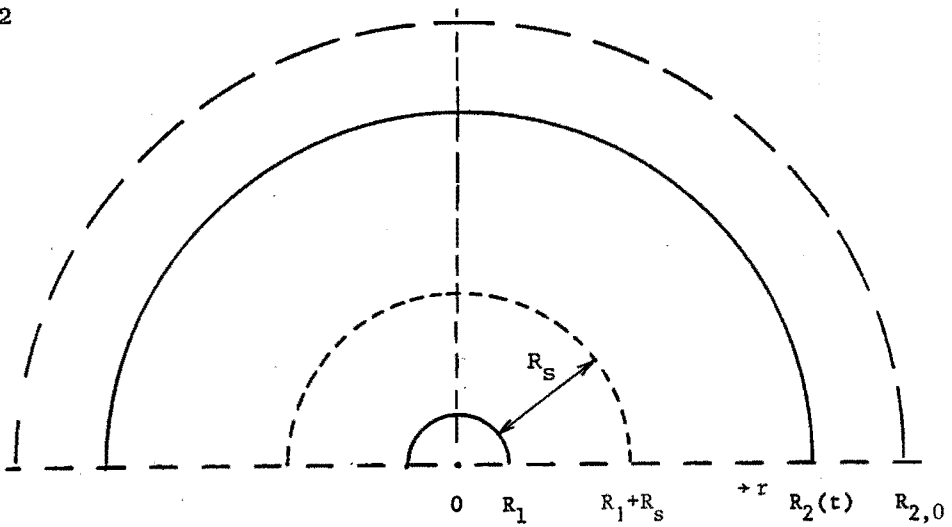
$$-D\rho_s \frac{\partial(\rho_m/\rho_s)}{\partial r} = j_{mi}^s(t) \quad (3.5)$$

in which  $\rho_m$  is the volume based concentration of the moisture ( $\text{kg}/\text{m}^3$ );  $t$  is time (s);  $r$  is the space coordinate (m);  $R_1$  is the fixed internal radius of a hollow cylinder or sphere (see Figure 3.1); in case of a one sided drying slab  $R_1=0$ ;  $R_2(t)$  is the (dynamic) external radius of the (shrinking) body at time  $t$ ;  $n_m$  is the moisture flux at space  $r$  ( $\text{kg}/\text{m}^2\text{s}$ ) with respect to stationary space coordinates;  $j_{mi}^s$  is the moisture flux through the external interface ( $\text{kg}/\text{m}^2\text{s}$ );  $v$  is the geometry parameter, with

$v = 0$  infinite slab

$v = 1$  infinite massive or hollow cylinder

$v = 2$  massive or hollow sphere



**Figure 3.1** Illustration of the boundary planes for  
(non-)shrinking hollow cylinders and spheres.

### 3.3 Dimensionless Diffusion Equation.

The mathematical transformation of the diffusion equation (eqns. 3.1-3.5) into a dimensionless form is described in Appendix A. Similar transformations for maximum- and non-shrinking systems have been given by Schoeber [23], Liou [28] and Van de Lijn [21]. In this thesis their work is generalized to massive and hollow systems with any degree of shrinkage. The transformation of the diffusion equation results in:

Generalized partial differential equation:

$$\frac{\partial m}{\partial \tau} = \frac{\partial}{\partial \phi} \left( D_r X^2 \frac{\partial m}{\partial \phi} \right) \quad (3.6)$$

Initial condition:

$$\tau = 0 \quad 0 \leq \phi \leq 1 \quad m = 1 \quad (3.7)$$

Boundary conditions:

$$\tau > 0 \quad \phi = 0 \quad X \frac{\partial m}{\partial \phi} = 0 \quad (3.8)$$

$$\phi = 1 \quad m = m_i \quad (3.9)$$

or

$$-D_r X_i \frac{\partial m}{\partial \phi} = F \quad (3.10)$$

The various (dimensionless) parameters and the shrinkage model used are explained in the following sections.

### 3.3.1 Mass Concentration.

The solids based moisture concentration  $u$  is defined as kg moisture per kg dry solid (kg m/kg ds), so that:

$$u = \frac{\rho_m}{\rho_s} \quad (3.11)$$

The dimensionless concentration  $m$  is defined as:

$$m = \frac{u - u_{\#}}{u_0 - u_{\#}} = \frac{v - v_{\#}}{v_0 - v_{\#}} \quad \text{with } 0 \leq m \leq 1 \quad (3.12)$$

in which  $v$  is the ratio of the volume fractions of moisture ( $\rho_m/d_m$ ) and solid ( $\rho_s/d_s$ ), expressed by:

$$v = \frac{d_s}{d_m} \frac{\rho_m}{\rho_s} = \frac{d_s}{d_m} u \quad (3.13)$$

In this expression  $d_s$  and  $d_m$  are the partial densities of solid and moisture respectively. It is assumed that the partial densities are independent of composition and are equal to the densities of the pure components ( $d_m = d_{mp}$ ,  $d_s = d_{sp}$ ). The initial value of  $v$  is denoted as  $v_0$ , the averaged value as  $\bar{v}$  and the equilibrium value as  $v_{*}$ ;  $v_{\#}$  is some arbitrary reference value, e.g. the value where the diffusion coefficient is practically zero, or the equilibrium value  $v_{*}$ .

### 3.3.2 Space Coordinate.

The dimensionless solids based space coordinate  $\phi$  is defined as:

$$\phi = \frac{\int_{R_1}^r \rho_s r^v dr}{\int_{R_1}^{R_2} \rho_s r^v dr} \quad \text{with } 0 \leq \phi \leq 1 \quad (3.14)$$

This space coordinate is based on a ratio of amounts of dry solids. Introduction of this space coordinate turns a dynamic external boundary ( $r=R_{2,t}$ ) into a fixed one ( $\phi=1$ ).

### 3.3.3 Diffusion Coefficient.

The dimensionless diffusion coefficient  $D_r$  is defined as:

$$D_r = \frac{D \rho_s^2}{D_0 \rho_{s0}^2} \quad (3.15)$$

in which  $D$  is the actual diffusion coefficient ( $m^2/s$ );  $D_0$  represents the value of the diffusion coefficient at  $\rho_{m0}$ .

### 3.3.4 Time.

The dimensionless time  $\tau$  is defined as:

$$\tau = \frac{D_0 \rho_{s0}^2 t}{(d_{s,ap} R_s)^2} \quad (3.16)$$

where the solid radius  $R_s$  is the thickness of a one sided drying slab or the thickness of the shell of a hollow body if all the water has been removed ( $\bar{v}=0$ );  $d_{s,ap}$  is the apparent density of the moisture free (porous) solid; it will be obvious that  $d_{s,ap}$  will depend on the degree of shrinkage and the initial moisture content. In general it is true that :

$$\begin{aligned} \rho_{s0} &\leq d_{s,ap} \leq d_{sp} \\ \text{and} \quad R_{s,p} &\leq R_s \leq R_{2,0} - R_1 \end{aligned} \quad (3.17)$$

### 3.3.5 Hollowness Factor.

The hollowness factor  $\lambda$  of the drying body is defined as:

$$\lambda = \frac{R_1}{R_1 + R_s} \quad \text{with } 0 \leq \lambda \leq 1 \quad (3.18)$$

By this definition the hollowness factor  $\lambda$  remains constant during drying. For massive bodies  $\lambda=0$ ; if  $\lambda \rightarrow 1$  the shell thickness of a hollow body is extremely thin with respect to its external radius, so in case of  $\lambda$ -values approaching 1 the hollow geometry tends to be similar to the slab geometry. For this reason the solids radius  $R_s$  of hollow systems is based on the dry shell thickness and not on the dry body radius, as is done by Schoeber [23].

### 3.3.6 Flux Parameter.

The dimensionless flux parameter  $F$  is defined as:

$$F = \frac{j_{mi}^s d_{s,ap} R_s}{D_0 \rho_{s0}^2 (u_0 - u_{\#})} \quad (3.19)$$

During drying the flux parameter  $F$  will only change if  $j_{mi}^s$  changes, because all other quantities in equation 3.19 are constants.

### 3.3.7 X-parameter.

The definition of the X-parameter is given by:

$$X = (v+1) \frac{1-\lambda}{1-\lambda^{v+1}} \left[ \lambda^{v+1} + (1-\lambda)^{v+1} \right] d_{s,ap} \int_0^{\phi} \frac{d\phi}{\rho_s} \Big|^{v+1} \quad (3.20)$$

and  $X_i$  follows by putting  $\phi=1$  in the above equation. The physical meaning of  $X_i$  will be explained in § 3.4.

The X-parameter still contains the volume based concentration  $\rho_s$ . The relation between  $\rho_s$  and  $\rho_m$  depends on the porosity of the material and therefore  $\rho_s$  can only be expressed in terms of the dimensionless concentration  $m$  if the shrinkage behaviour is known (see § 3.3.8).

### 3.3.8 Shrinkage Model.

Quite often the volume  $V$  of a material is a linear function of the average moisture content [44], which is expressed by:

$$V = V_s (1 + \sigma \bar{v}) \quad (3.21)$$



where the solids volume  $V_s$  is the moisture free volume of the material;  $\bar{v}$  is the ratio of the averaged volume fractions of moisture and solid (eqn. 3.13);  $\sigma$  is the *shrinkage coefficient*, which is actually defined by equation 3.21.

The solids mass balance reads:

$$\bar{\rho}_s V = d_{s,ap} V_s \quad (3.22)$$

so from equations 3.21 and 3.22 follows:

$$\frac{d_{s,ap}}{\bar{\rho}_s} = 1 + \sigma \bar{v} \quad (3.23)$$

For non-shrinking systems  $\bar{\rho}_s = \rho_{s0} = d_{s,ap}$ , so  $\sigma = 0$ .

For maximum-shrinking systems the volume balance reads:

$$\frac{\bar{\rho}_m}{d_m} + \frac{\bar{\rho}_s}{d_s} = 1 \quad (3.24)$$

or

$$\frac{d_s}{\bar{\rho}_s} = 1 + \bar{v} \quad (3.25)$$

Because now  $d_s = d_{sp} = d_{s,ap}$  it follows that  $\sigma = 1$ .

In general, for drying bodies with constant partial quantities the shrinkage coefficient will obey to  $0 \leq \sigma \leq 1$ .

Assuming that shrinkage occurs isotropic and homogeneous and that  $\sigma$  is independent of moisture content, equation 3.23 will also be valid for local concentrations. Therefore:

$$\frac{d s_{a,p}}{\rho_s} = 1 + \sigma v \quad (3.26)$$

and the expression for the  $X$ -parameter now becomes:

$$X = (v+1) \frac{1-\lambda}{1-\lambda^{v+1}} \left[ \lambda^{v+1} + (1-\lambda)^{v+1} \int_0^{\phi} (1+\sigma v) d\phi \right]^{\frac{v}{v+1}} \quad (3.27)$$

where  $v$  can be expressed in terms of the dimensionless concentration  $m$  by (eqn. 3.12):

$$v = v_{\#} + (v_0 - v_{\#})m \quad (3.28)$$

The  $X_i$ -parameter follows from equation 3.27 with  $\phi=1$ :

$$X_i = X_{i,\sigma=0} \left[ 1 + (1-\lambda)^{v+1} \sigma v \right]^{\frac{v}{v+1}} \quad (3.29)$$

where:

$$X_{i,\sigma=0} = (v+1) \frac{1-\lambda}{1-\lambda^{v+1}} \quad (3.30)$$

### 3.3.9 Generalized Biot Number.

$j_{mi}^s$  in equation 3.19 must also obey the mass transfer equation 2.41 in the gas phase:

$$j_{mi}^s = k_f' \rho_f' \ln \left[ \frac{1-\omega_{mi}^{\infty}}{1-\omega_{mi}'} \right] \quad (3.31)$$

The moisture activity  $a_m$  for ideal gas mixtures is defined as:

$$a_m = \frac{P_m}{P_{m,sat}} = \frac{\rho_m'}{\rho_{m,sat}'} \quad (3.32)$$

where  $P_m$  is the partial pressure ( $N/m^2$ ) of the moisture in the gas phase,  $P_{m,sat}$  is the saturation pressure of the moisture at the given temperatures;  $\rho_m'$  and  $\rho_{m,sat}'$  are moisture concentrations ( $kg/m^3$ ) in the gas phase.

The mass transfer coefficient ( $k''$ ), based on a difference in moisture activities, is defined by:

$$j_{mi}^s = k''(a_{mi} - a_{m\infty}) \quad (3.33)$$

From equations 3.31 and 3.33 it can be derived that for constant  $k''$ -values the value of  $k''$  will remain constant only if the temperature remains constant.

Substitution of expression 3.33 into equation 3.14 yields:

$$F = Bi \cdot m_i \quad (3.34)$$

where  $Bi$  is the generalized Biot number, defined as:

$$Bi = \frac{\gamma k'' d_{s,ap} R_s}{D_0 \rho_{s0}^2} \quad (3.35)$$

in which  $\gamma$  follows from the equilibrium relation  $a_m$  versus  $u$  (sorption isotherm; see Figure 4.1):

$$\gamma = \frac{a_{mi} - a_{m\infty}}{u_i - u_{*x}} \quad (3.36)$$

Substitution of equation 3.34 into the external boundary condition

3.10 yields the most general formulation of the external boundary condition. In chapter IV it will be elucidated that this condition sometimes may be formulated as a constant surface flux or as a constant surface concentration.

### 3.4 Mass Balance and Drying Time.

The moisture balance over the drying body during a time interval  $dt$  reads:

$$j_{mi}^s A dt = -\bar{\rho}_s V d(\bar{\rho}_m / \bar{\rho}_s) \quad (3.37)$$

where  $A$  is the mass exchanging area and  $V$  is the (shell-)volume of the drying body.

The averaged drying efficiency  $E$  is defined as the fraction of moisture removed:

$$E = \frac{v_0 - \bar{v}}{v_0 - v_{\#}} = \frac{u_0 - \bar{u}}{u_0 - u_{\#}} = 1 - \bar{m} \quad (3.38)$$

Putting the balance in a dimensionless form one finds:

$$FX_1 d\tau = dE \quad (3.39)$$

where

$$X_1 = \frac{A d_{s,ap} R_s}{\bar{\rho}_s V} = \frac{A R_s}{V_s} \quad (3.40)$$

It can be proven that  $X_1$  according to equation 3.40 equals  $X_i$

according to equation 3.29. However, equation 3.40 offers a better insight in the physical meaning of  $X_i$ . One can visualize  $X_i$  as the ratio of the solids volumina of a slab and the actual body, both with the same external area  $A$  and the same solids radius  $R_s$ . Thus for slabs it simply follows that  $X_i=1$ , which is in agreement with equation 3.29. Further, during a drying process  $X_i$  will only change if the surface area  $A$  changes, because  $R_s$  and  $V_s$  will remain constant. In this view  $X_i$  may be looked upon as a dimensionless surface area.

Integration of equation 3.39 gives the basic equation for the calculation of the drying time:

$$\tau = \int_0^E \frac{dE}{FX_i} \quad (3.41)$$

It is obvious that the drying time can be calculated if both  $F$  and  $X_i$  are known as functions of  $E$ .

The relationship between  $X_i$  and  $E$  is given by equations 3.29, and 3.38. These equations can be rearranged to:

$$X_i = X_{i0} (1-sE)^{\frac{v}{v+1}} \quad (3.42)$$

in which

$$X_{i0} = X_{i,\sigma=0} \left[ 1 + (1-\lambda^{v+1})\sigma v_0 \right]^{\frac{v}{v+1}} \quad (3.43)$$

and

$$s = \frac{(1-\lambda^{v+1})\sigma(v_0 - v_{\#})}{1 + (1-\lambda^{v+1})\sigma v_0} \quad (3.44)$$

The parameter  $s$  is a *modified shrinkage coefficient*, which indicates to what extent the shrinking properties of the material will influence the drying behaviour, e.g. a material with a low initial water content  $v_0$  will hardly shrink any more even if  $\sigma$  has a high value.

The relationship between  $F$  and  $E$  is far more complicated to be found and the following chapters of this thesis are dealing with this problem:

Chapter IV: non-shrinking systems ( $\sigma=0$ ) with a concentration independent diffusion coefficient ( $D_r=1$ ).

Chapter V: (non-)shrinking systems ( $0 \leq \sigma \leq 1$ ) with a power law dependence of the diffusion coefficient ( $D_r = m^a$ ).

### 3.5 Summary of Generalized Diffusion Equation, Mass Balance and Definitions of Dimensionless Parameters.

Generalized partial differential equation:

$$\frac{\partial m}{\partial \tau} = \frac{\partial}{\partial \phi} (D_r X^2 \frac{\partial m}{\partial \phi})$$

Initial condition:

$$\tau = 0 \quad 0 \leq \phi \leq 1 \quad m = 1$$

Boundary conditions:

$$\tau > 0 \quad \begin{array}{l} \phi = 0 \\ \phi = 1 \end{array} \quad \begin{array}{l} X \frac{\partial m}{\partial \phi} = 0 \\ m = m_i \end{array}$$

or

$$-D_r X_i \frac{\partial m}{\partial \phi} = F$$

Mass balance:

$$FX_i d\tau = dE \quad \longrightarrow \quad \tau = \int_0^E \frac{dE}{FX_i}$$

$$X_i = X_{i0} (1-sE)^{\frac{\nu}{\nu+1}}$$

$$X_{i0} = X_{i,\sigma=0} \left[ 1 + (1-\lambda)^{\nu+1} \sigma v_0 \right]^{\frac{\nu}{\nu+1}}$$

$$X_{i,\sigma=0} = \frac{(v+1)^{1-\lambda}}{1-\lambda^{\nu+1}}$$

$$s = \frac{(1-\lambda)^{\nu+1} \sigma (v_0 - v_{\#})}{1 + (1-\lambda)^{\nu+1} \sigma v_0}$$

Summary of definitions:

concentration: $u = \frac{\rho_m}{\rho_s}$	diffusion coefficient: $D_r = \frac{D \rho_s^2}{D_0 \rho_{s0}}$
concentration: $v = \frac{d_s}{d_m} \frac{\rho_m}{\rho_s} = \frac{d_s}{d_m} u$	time: $\tau = \frac{D_0 \rho_{s0}^2 t}{(d_{s,ap} R_s)^2}$
concentration: $m = \frac{u - u_{\#}}{u_0 - u_{\#}} = \frac{v - v_{\#}}{v_0 - v_{\#}}$	averaged efficiency: $E = \frac{v_0 - \bar{v}}{v_0 - v_{\#}} = 1 - \bar{m}$
space coordinate: $\phi = \frac{\int_{R_1}^r \rho_s r^{\nu} dr}{\int_{R_1}^{R_2} \rho_s r^{\nu} dr}$	flux parameter: $F = \frac{j_{mi}^s d_{s,ap} R_s}{D_0 \rho_{s0}^2 (u_0 - u_{\#})}$
hollowness factor: $\lambda = \frac{R_1}{R_1 + R_s}$	Biot number: $Bi = \frac{\gamma k'' d_{s,ap} R_s}{D_0 \rho_{s0}^2}$
$X = (v+1) \frac{1-\lambda}{1-\lambda^{\nu+1}} \left[ \lambda^{\nu+1} + (1-\lambda)^{\nu+1} \int_0^{\phi} (1+\sigma v) d\phi \right]^{\frac{\nu}{\nu+1}}$	





CHAPTER IVNON-SHRINKING SYSTEMS WITH CONCENTRATION INDEPENDENT DIFFUSIONCOEFFICIENT4.1 Diffusion Equation in Linear  $r$ -Coordinates.

For isothermal drying of non-shrinking bodies with concentration independent diffusion coefficient analytical solutions of the diffusion equation can be obtained. In order to find these solutions we depart from the dimensional diffusion equation (§3.2), which in a form adapted for this case reads:

Partial differential equation:

$$\frac{\partial \rho_m}{\partial t} = \frac{1}{r^v} \frac{\partial}{\partial r} (r^v D \frac{\partial \rho_m}{\partial r}) \quad (4.1)$$

Initial condition:

$$t = 0 \quad R_1 < r < R_2 \quad \rho_m = \rho_{m0} \quad (4.2)$$

Boundary conditions:

$$t > 0 \quad r = R_1 \quad \frac{\partial \rho_m}{\partial r} = 0 \quad (4.3)$$

$$r = R_2 \quad \rho_m = \rho_{mi} \quad (4.4)$$

or

$$-D \frac{\partial \rho_m}{\partial r} = j_{mi}^s \quad (4.5)$$

**Table 4.1** Summary of parameters for non-shrinking system.

concentration:	$m = \frac{\rho_m - \rho_{m^*}}{\rho_{m0} - \rho_{m^*}}$	(4.6)
space	$\phi = \frac{r^{\nu+1} - R_1^{\nu+1}}{R_2^{\nu+1} - R_1^{\nu+1}}$	(4.7)
diffusion coefficient	$D_r = \frac{D}{D_0}$	(4.8)
time	$\tau = \frac{D_0 t}{(R_2 - R_1)^2}$	(4.9)
hollowness factor	$\lambda = \frac{R_1}{R_2}$	(4.10)
flux parameter	$F = \frac{j_{m1}^s (R_2 - R_1)}{D_0 (\rho_{m0} - \rho_{m^*})}$	(4.11)
Biot number	$Bi = \frac{\gamma}{\rho_{s0}} \frac{k'' (R_2 - R_1)}{D_0}$	(4.12)
local efficiency	$E' = \frac{\rho_{m0} - \rho_m}{\rho_{m0} - \rho_{m^*}} = 1 - m$	(4.13)
average efficiency	$E = \frac{\rho_{m0} - \bar{\rho}_m}{\rho_{m0} - \rho_{m^*}} = 1 - \bar{m}$	(4.14)
	$X = X_{i,\sigma=0} \left[ \lambda^{\nu+1} + (1-\lambda)^{\nu+1} \phi \right]^{\frac{\nu+1}{\nu}}$	(4.15)
	$X_i = \frac{A(R_2 - R_1)}{V} = X_{i,\sigma=0} = (\nu+1) \frac{1-\lambda}{1-\lambda^{\nu+1}}$	(4.16)

Non-shrinking systems are characterized by  $\sigma=0$  and  $\rho_s = \rho_{s0} = d_{s,ap}$ .

Choosing the equilibrium concentration as reference value ( $m_{\#} = m_{*}$ ), the dimensionless parameters as defined in Chapter III now become as given in Table 4.1.

In the literature no solutions are found in terms of a volume based space coordinate ( $\Phi$ ), therefore a dimensionless linear space coordinate ( $y$ ) is introduced:

$$y = \frac{r - R_1}{R_2 - R_1} \quad (4.17)$$

Substitution of the dimensionless parameters into the equations 4.1-4.5 gives, in case of a constant diffusion coefficient ( $D=D_0$ , so  $D_r=1$ ), the following diffusion equation in linear  $y$ -coordinates:

Partial differential equation:

$$\frac{\partial m}{\partial \tau} = \frac{1}{[\lambda + (1-\lambda)y]^v} \frac{\partial}{\partial y} ([\lambda + (1-\lambda)y]^v \frac{\partial m}{\partial y}) \quad (4.18)$$

Initial condition:

$$\tau = 0 \quad 0 < y < 1 \quad m = 1 \quad (4.19)$$

Boundary conditions:

$$\tau > 0 \quad y = 0 \quad \frac{\partial m}{\partial y} = 0 \quad (4.20)$$

$$y = 1 \quad m = m_i \quad (4.21)$$

or

$$-\frac{\partial m}{\partial y} = F \quad (4.22)$$

From the analytical solutions of the above differential equation (implicit) expressions for the drying time  $\tau$  as a function of the average efficiency  $E$  can be derived. In the short-cut approach, as

will be described in this chapter. (explicit) expressions for  $\tau$  versus  $E$  are obtained from the integration of the mass balance:

$$\tau = \frac{1}{X_{i,\sigma=0}} \int_0^E \frac{dE}{F} \quad (4.23)$$

The integration requires the relationship between fluxparameter  $F$  and average efficiency  $E$ . Finding approximate equations for this relationship, which appears to depend on the drying stage, is the main theme of this chapter.

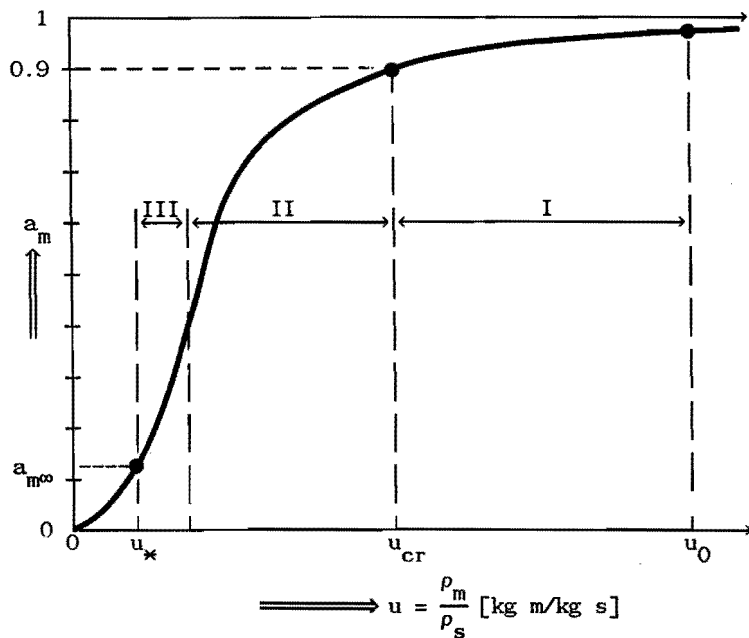
#### 4.2 Drying Stages.

Let us assume an adiabatic drying process with constant external conditions, which means constant temperature  $\theta'_\infty$ , constant humidity  $\omega'_{m\infty}$ , and constant mass transfer coefficient  $k'$ . Further, assume that the initial concentration  $u_0$  of the material is such, that the corresponding equilibrium moisture activity  $a_m$  approaches 1 (see Figure 4.1). Thus, after a relatively short time the material will take on the wet bulb temperature. For a drying system one can safely assume that at the interface the thermodynamic equilibrium relation  $a_m$  versus  $u$  (sorption-isotherm) holds. Thus the local values at the interface ( $u_i, a_{mi}$ ) can be obtained from Figure 4.1.

The moisture flux  $j_{mi}^S$  through the interface, expressed in terms of gas phase conditions (§2.3.2), reads:

$$j_{mi}^S = k' \rho_f' \ln \left[ \frac{1 - \omega'_{m\infty}}{1 - \omega'_{mi}} \right] \quad (4.24)$$

As long as the surface moisture activity  $a_{mi}$  remains nearly constant



**Figure 4.1**

Schematic representation of a sorption-isotherm and some characteristic quantities (for explanation see text).

**Main Drying Stages:**

- I : nearly constant surface moisture activity  
(constant boundary flux for non-shrinking systems)
- II : decreasing surface moisture activity
- III : nearly constant surface moisture concentration  
(constant boundary concentration)

(e.g.  $a_{mi} > 0.9$ ), then  $\omega'_{mi}$  will remain nearly constant (see eqn. 3.32) and therefore the boundary flux  $j_{mi}^s$  will remain nearly constant as well.

This first drying stage (I), often referred to as the "constant flux period" or "constant activity period", is fully controlled by external conditions: the drying material behaves almost like pure moisture. For the boundary condition of the diffusion equation at  $y=1$  (eqn. 4.22) now holds:

$$F = F_{ca} \approx \text{constant}$$

The end of the constant activity period is arbitrarily defined at  $a_{mi} = 0.9$ . At this moment the surface moisture concentration has reached the so called critical value  $u_{cr}$ . From now on the surface moisture activity starts to decrease significantly and drying stage II has started. It will be obvious from equation 4.24 that the boundary flux will decrease. Consequently, the temperature of the drying material gradually increases towards the dry-bulb temperature. The boundary condition of the diffusion equation at  $y=1$  is now given by equation 4.22, with:

$$F = Bi \cdot m_i$$

For a non-shrinking system the Biot number is given by equation 4.12; the value of  $\gamma$  follows from the sorption isotherm. If, in the concentration interval of interest, the sorption-isotherm may be approximated by a linear relationship ( $\gamma \approx \text{constant}$ ) and  $k''$  does not change too much despite of an increasing gas film temperature, then the Biot number will remain nearly constant during drying. However, in most practical drying situations this will not be the case.

Arbitrarily, drying stage II ends and drying stage III starts, when the surface concentration reaches a nearly constant value (e.g.  $m_i \leq 0.1$ ). During this final drying stage, often referred to as

"constant boundary concentration period" or "zero boundary concentration period", the relative changes in the surface concentration  $u_i$  with respect to the maximum attainable change ( $u_0 - u_{*}$ ) remain small. The boundary condition for the diffusion equation is now best given by equation 4.21, with:  $m_i \approx 0$ .

During this final drying stage the temperature of the material gradually increases until, in case of complete equilibrium, the dry-bulb temperature has been reached.

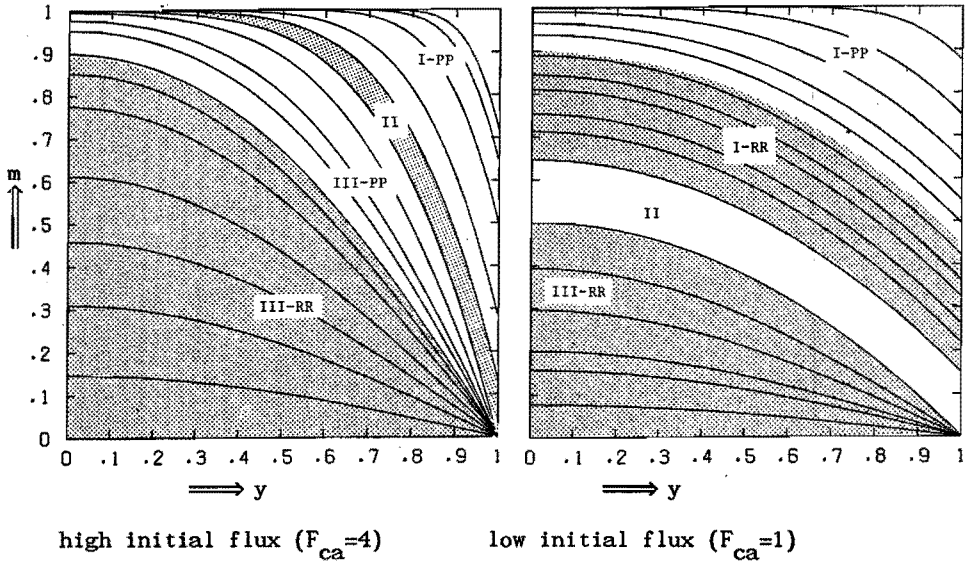
Each of the three main stages can be split up into two substages, namely:

- *Penetration Period*, during which the centre concentration changes hardly from the initial value (e.g.  $m_{\text{centre}} \geq 0.9$ ).
- *Regular Regime*, during which the centre concentration changes significantly from its initial value (e.g.  $m_{\text{centre}} \leq 0.9$ ).

Not all six possible drying stages can occur during a drying process. The occurrence of drying stages depends on the initial conditions (especially the starting flux) and the physical properties of the material (especially the sorption-isotherm). In Figure 4.2 two examples are given to illustrate the subsequent occurrence of stages during a drying process.

This thesis only deals with the drying stages I (nearly constant surface activity) and III (nearly constant surface concentration); drying stage II (decreasing surface activity) was not investigated (see §1.4).

As a first physical approximation a constant surface activity for stage I and a constant surface concentration for stage III are assumed in the remaining part of this thesis.



**Figure 4.2** Two examples of the subsequent occurrence of drying stages for a non-shrinking slab with a constant diffusion coefficient (PP=Penetration Period; RR=Regular Regime).

#### 4.3 Drying Stage with Constant Surface Flux.

The end of this drying stage is defined as the moment, where the local drying efficiency at the interface has reached the critical value

$E_{i,cr}^*$ ; the corresponding average critical efficiency is denoted  $E_{cr}$ .

The drying time follows from the integrated mass balance:

$$\tau = \frac{E}{F_{ca} X_{i,\sigma=0}} \quad (4.25)$$

The duration of the constant activity period  $\tau_{ca}$  follows from equation

4.25, if  $E_{cr}$  is known. Values of  $E_{cr}$  decrease with increasing values

of the constant flux parameter  $F_{ca}$ . The relationship between  $E_{cr}$  and



$F_{ca}$ , the so-called "critical-point curve", depends on the critical surface concentration  $E'_{i,cr}$ . The value of  $E'_{i,cr}$  is derived from the sorption-isotherm; next the critical-point curve is known from the relationship among  $E$ ,  $E'_i$  and  $F_{ca}$ ; for this the diffusion equation has to be solved.

#### 4.3.1 Analytical Solutions ( $F_{ca} = \text{constant}$ ).

The solutions for the massive systems and hollow cylinders can be found in literature [37-39]; the diffusion equation of the hollow sphere was solved by Bosch [40]. Here the solutions are given in two different ways: one obtained by solving the diffusion equation via separation of variables and the other obtained via Laplace transformation. In Appendix B the solutions for massive and hollow systems, obtained by separation of variables, are given. In this main text only the solutions for massive systems will be presented.

#### Slab ( $v=0$ )

Via separation of variables:

$$\frac{E'}{F_{ca}} = \tau + \frac{1}{2}y^2 - \frac{1}{6} - 2 \sum_1^{\infty} \frac{\cos(\mu_k y)}{\cos(\mu_k)} \frac{\exp(-\mu_k^2 \tau)}{\mu_k^2} \quad (4.26)$$

in which  $\mu_k$  are the positive roots of the characteristic equation :  $\sin(\mu_k) = 0$ , thus  $\mu_k = k\pi$  and  $\cos(\mu_k) = (-1)^k$ .

Via Laplace transformation:

$$\frac{E'}{F_{ca}} = 2 \frac{\sqrt{\tau}}{\sqrt{\pi}} \sum_0^{\infty} \left[ \exp\left(\frac{-(2k+1-y)^2}{4\tau}\right) - \frac{2k+1-y}{2\sqrt{\tau}} \sqrt{\pi} \operatorname{erfc}\left(\frac{2k+1-y}{2\sqrt{\tau}}\right) + \exp\left(\frac{-(2k+1+y)^2}{4\tau}\right) - \frac{2k+1+y}{2\sqrt{\tau}} \sqrt{\pi} \operatorname{erfc}\left(\frac{2k+1+y}{2\sqrt{\tau}}\right) \right] \quad (4.27)$$

Massive cylinder ( $\nu=1$ )

Via separation of variables:

$$\frac{E'}{F_{ca}} = 2\tau + \frac{1}{2}y^2 - \frac{1}{4} - 2 \sum_1^{\infty} \frac{J_0(\mu_k y)}{J_0(\mu_k)} \frac{\exp(-\mu_k^2 \tau)}{\mu_k^2} \quad (4.28)$$

in which  $\mu_k$  are the positive roots of the characteristic equation :

$J_1(\mu_k) = 0$ ;  $J_0$  and  $J_1$  are Bessel functions [41] of the first kind of order 0 respectively order 1.

Via Laplace transformation:

$$\begin{aligned} \frac{E'}{F_{ca}} = & 2 \frac{\sqrt{\tau}}{\sqrt{y}} \operatorname{ierfc}\left(\frac{1-y}{2\sqrt{\tau}}\right) + \frac{1+3y}{2y} \frac{\tau}{\sqrt{y}} i^2 \operatorname{erfc}\left(\frac{1-y}{2\sqrt{\tau}}\right) + \\ & \frac{9+33y^2+6y}{16y} \frac{\tau\sqrt{\tau}}{\sqrt{y}} i^3 \operatorname{erfc}\left(\frac{1-y}{2\sqrt{\tau}}\right) + \dots \end{aligned} \quad (4.29)$$

Massive sphere ( $\nu=2$ )

Via separation of variables :

$$\frac{E'}{F_{ca}} = 3\tau + \frac{1}{2}y^2 - \frac{3}{10} - \frac{2}{y} \sum_1^{\infty} \frac{\sin(\mu_k y)}{\sin(\mu_k)} \frac{\exp(-\mu_k^2 \tau)}{\mu_k^2} \quad (4.30)$$

in which  $\mu_k$  are the positive roots of the characteristic equation :

$$\tan(\mu_k) = \mu_k.$$

Via Laplace transformation:

$$\frac{E'}{F_{ca}} \approx \frac{1}{y} \left[ \exp[-(1-y)\tau] \operatorname{erfc}\left(\frac{1-y}{2\sqrt{\tau}} - \sqrt{\tau}\right) - \operatorname{erfc}\left(\frac{1-y}{2\sqrt{\tau}}\right) \right] \quad (4.31)$$

This latter solution is an approximation. The exact solution can not be found, because of an unsolved back transformation from the Laplace

domain [40]. However, the given approximation appears to be quite close to the exact solution, obtained via separation of variables (eqn. 4.30).

#### 4.3.2 Penetration Period ( $F_{ca} = \text{constant}$ ).

At sufficiently small  $\tau$  values the concentration profiles are not yet penetrated into the centre of the system, in other words : the centre concentration has hardly changed from its initial value. During this period, which is called "*Penetration Period with constant boundary flux*", the concentration profiles are independent of the hollowness factor  $\lambda$ .

The solutions, obtained via Laplace transformation, reduce for low  $\tau$  values to the first term of the series. Nevertheless, a fairly complicated expression remains, which can be further simplified by developing the exponential- and error functions in infinite series [41], which in their turn reduce to a few dominant terms for small  $\tau$  values. The following expression appears to be a very good approximation for the surface efficiency (maximum deviations about 2% for  $\tau \leq 0.15$ ):

$$\frac{E'_i}{F_{ca}} = \frac{2}{\sqrt{\pi}} \sqrt{\tau} + \frac{\nu}{2}(1-\lambda)\tau + \frac{\nu(\nu+2)}{6\sqrt{\pi}}(1-\lambda)^2 \tau \sqrt{\tau} + \frac{\nu(\nu+2)}{16}(1-\lambda)^3 \tau^2 \quad (4.32)$$

For extremely small  $\tau$  values, equation 4.32 reduces to:

$$\frac{E'_i}{F_{ca}} = \frac{2}{\sqrt{\pi}} \sqrt{\tau} \quad (4.33)$$

Thus, for this extreme situation  $E'_i/F_{ca}$  only depends on  $\tau$  and is independent of the geometry parameter  $\nu$  and the hollowness factor  $\lambda$ ;

in this respect all massive and hollow systems behave like a slab. Elimination of  $\tau$  from equation 4.33 by means of the mass balance (eqn. 4.25) gives :

$$\frac{F_{ca}}{E'_i} = \frac{\pi}{4} X_{i,\sigma=0} \frac{E'_i}{E} \quad (4.34)$$

We now define a G-parameter:

$$G = \frac{\frac{F}{E'_i} \frac{E}{E'_i}}{X_i} \quad (4.35)$$

From equations 4.34, 4.35 and 4.36 follows the initial value  $G_0$ :

$$G_0 = \lim_{\tau \rightarrow 0} G = \lim_{E \rightarrow 0} G = \frac{\pi}{4} \quad (4.36)$$

It can be concluded that the  $G_0$ -parameter is independent of geometry and hollowness, in other words: the  $G_0$  parameter may be looked upon as a quantity expressing that all hollow and massive geometries behave like a slab geometry at  $\tau \rightarrow 0$ .

For small  $\tau$ -values, elimination of  $\tau$  from equation 4.32 and the mass balance (eqn. 4.25), yields a rather complicated expression. A better approach is based on finding a correlation of the exact calculated values of  $F_{ca}/E'_i$  versus  $E'_i/E$ , which appears to be a nearly straight line :

$$\frac{F_{ca}}{E'_i} = G_0 X_{i,\sigma=0} \left[ \frac{E'_i}{E} - \alpha \right] \quad (4.37)$$

in which for the correlation parameter  $\alpha$  the following relations can be obtained by fitting:

$$\text{For a slab} \quad : \quad \alpha = 0 \quad (4.38a)$$

$$\text{For a cylinder: } \alpha = 0.60 - 0.12\lambda - 0.31\lambda^2 - 0.17\lambda^3 \quad (4.38b)$$

$$\text{For a sphere} \quad : \quad \alpha = 0.76 - 0.08\lambda + 0.04\lambda^2 - 0.72\lambda^3 \quad (4.38c)$$

Note that equation 4.37 can also be expressed as:

$$G = G_0 \left(1 - \alpha \frac{E}{E_i'}\right) \quad (4.39)$$

and  $\alpha$  can also be found by putting  $G$  versus  $E/E_i'$ .

#### 4.3.3 Regular Regime ( $F_{ca} = \text{constant}$ ).

At sufficiently high  $\tau$  values, in case of low drying rates, the solutions obtained via separation of variables degenerate to a very simple form, because all terms in the series are negligible small compared with  $\tau$ . During this drying stage concentration profiles are parabolic and decrease linearly with time (see Figure 4.2).

Irrespective of the values of the constant surface flux and the initial moisture content, eventually the concentration profiles will take on the parabolic shape. This drying stage is called "**Regular Regime with constant boundary flux**".

From equations 4.26, 4.28 and 4.30 the following expression for the surface efficiency can be derived:

$$\frac{E_i'}{F_{ca}} = (v+1)\tau + \frac{1}{v+3} \quad (4.40)$$

Elimination of  $\tau$  by using the mass balance (eqn. 4.25) gives:

$$F_{ca} = (v+3)(E'_i - E) \quad (4.41)$$

Defining the mass transfer coefficient  $k_d$  for the dispersed phase as follows:

$$j_{mi}^s = k_d(\bar{\rho}_m - \rho_{mi}) \quad (4.42)$$

or in a dimensionless form:

$$F = \frac{k_d(R_2 - R_1)}{D_0} (\bar{m} - m_i) \quad (4.43)$$

and defining the Sherwood number for the dispersed phase according to:

$$Sh_d = \frac{2k_d(R_2 - R_1)}{\bar{D}} \quad (4.44)$$

gives :

$$F = \frac{1}{2} Sh_d \bar{D}_r (\bar{m} - m_i) \quad (4.45)$$

where  $\bar{D}_r$  is an average reduced diffusion coefficient, defined as:

$$\bar{D}_r = \frac{\bar{D}}{D_0} = \frac{\int_{m_i}^{\bar{m}} D_r dm}{\bar{m} - m_i} \quad (4.46)$$

For concentration independent diffusion coefficient  $\bar{D}_r = D_r = 1$  equation

4.45 can be written as:

$$F = \frac{1}{2} \text{Sh}_d (E'_i - E) \quad (4.47)$$

For massive geometries the following simple expression for  $\text{Sh}_d$  can be derived from equations 4.41 and 4.47:

$$\text{Sh}_d = 6 + 2v \quad (4.48)$$

In a similar way the following expressions for  $\text{Sh}_d$  can be obtained in case of hollow cylinders:

$$\text{Sh}_d = 8 \frac{(1+\lambda^2)(1-\lambda)^3}{(1-4\lambda^2) + [3-4\ln(\lambda)]\lambda^4} \quad (4.49)$$

and for hollow spheres:

$$\text{Sh}_d = 10 \frac{(1+\lambda+\lambda^2)^2}{1+3\lambda+6\lambda^2+5\lambda^3} \quad (4.50)$$

Thus, during the Regular Regime with constant boundary flux  $\text{Sh}_d$  takes on constant values (eqns. 4.48, 4.49 and 4.50).

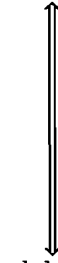
In Table 4.2 values of  $\text{Sh}_d$  are summarized. For a hollow geometry the values lay between those of the massive geometry ( $\lambda=0$ ) and the slab geometry ( $\lambda=1$ ).

It appears that equations 4.48-4.50 can be combined into one single handy relation:

$$\text{Sh}_d = 6 + 2v \left[ \frac{X_{1,\sigma=0}^{-1}}{v} \right]^{2.15-0.18v} \quad (4.51)$$

Only the exponent in this equation is a correlation parameter, obtained by fitting of the exact values. Deviations between approximate and exact values are less than 0.3% (see Table 4.2).

**Table 4.2:** Values of  $Sh_d$  during Regular Regime with constant boundary flux ( $D_r=1, \sigma=0$ ).

$\lambda$	cylinder ( $v=1$ )		sphere ( $v=2$ )		
	exact (eqn.4.49)	approx. (eqn.4.51)	exact (eqn.4.50)	approx. (eqn.4.51)	
0	8	8	10	10	massive  slab ( $v=0$ )
0.05	7.638	7.642	9.504	9.480	
0.1	7.341	7.347	9.026	8.999	
0.2	6.898	6.890	8.179	8.165	
0.3	6.594	6.591	7.503	7.504	
0.4	6.383	6.377	6.993	7.002	
0.5	6.236	6.230	6.622	6.633	
0.6	6.136	6.130	6.360	6.372	
0.7	6.069	6.066	6.185	6.195	
0.8	6.028	6.026	6.075	6.083	
0.9	6.006	6.006	6.017	6.021	
1	6	6	6	6	

#### 4.3.4 Transition from Penetration Period to Regular Regime

( $F_{ca} = \text{constant}$ ).

At high values of  $F_{ca}/E_i'$  the Regular Regime correlation (eqn. 4.47) gives too low values of the drying time; at low values of  $F_{ca}/E_i'$  the Penetration Period correlation (eqn. 4.37) predicts too high values of the drying time. The transition (T) is chosen nearby the value of  $F_{ca}/E_i'$  where the difference between the drying times, obtained from the two correlations, is minimal. Based on the exact solutions of the differential equation, it appears that the transition criterium can be formulated as:

$$\left[ \frac{F_{ca}}{E_i'} \right]_T = 1.5 - \lambda + \lambda^2 \quad (4.52)$$

For values of  $F_{ca}/E_i'$  larger than this criterium the correlation for the Penetration Period should be used, whereas at lower values the



correlation for the Regular Regime should be applied. Thus, the two drying periods are short-cut in a simple way: stepping over from the one correlation to the other at a specific value of  $F_{ca}/E'_i$ . The maximum error in the predicted drying time occurs at this transition point. By allowing maximum errors of about 4% the transition criterium could be formulated independent of the geometry parameter  $v$ .

Because initially  $E'_i=0$  and thus  $F_{ca}/E'_i \rightarrow \infty$ , every drying process with a constant boundary flux will show a Penetration Period. However, at sufficiently high drying fluxes the surface efficiency will reach the critical value  $E'_{i,cr}$  before the concentration profiles are penetrated into the centre of the material; from the transition criterium follows that this will happen if:

$$F_{ca} \geq (1.5 - \lambda + \lambda^2) E'_{i,cr} \quad (4.53)$$

*In these cases no Regular Regime with constant boundary flux will succeed the Penetration Period (see Figure 4.2).*

#### 4.3.5 Algorithm for Short-Cut Calculation ( $F_{ca} = \text{constant}$ ).

An algorithm for calculating  $\tau$  and E from given values of  $\nu$ ,  $\lambda$ ,  $F_{ca}$  and  $E_i'$  could take on the following form:

```

begin {short-cut algorithm,  $D_r=1$ ,  $\sigma=0$ ,  $F=F_{ca}$ }
read( $\nu, \lambda, F_{ca}, E_i'$ )
if  $\frac{F_{ca}}{E_i'} \geq 1.5 - \lambda + \lambda^2$ 
  then {Penetration Period}
    calculate  $\alpha$  from equations 4.38a-4.38c
    calculate E from equation 4.37
  else {Regular Regime}
    calculate  $Sh_d$  from equation 4.51
    calculate E from equation 4.47
calculate  $\tau$  from equation 4.25
write( $\tau, E$ )
end.
```

Comparing this short-cut calculation of  $\tau$  and E with the exact calculation method, reveals that the *maximum errors* in  $\tau$  and E occur near the transition point and are *less than 4%!!*

#### 4.4 Drying Stage with Constant Surface Concentration.

Assume a sufficiently high initial flux, so that the moisture concentration profiles hardly have penetrated into the drying material at reaching the condition of nearly constant surface concentration (stage III). Consequently, the moisture distribution in the material still will be nearly uniform and the drying time of the preceding

stages I and II will be negligible with respect to the total drying time of the material. During drying stage III the drying flux will decrease continuously and in case of an isothermal process the drying time should be calculated using equation 4.23 . The relationship between the flux  $F$  and the efficiency  $E$  has to be obtained from the (analytical) solution of the diffusion equation. For the situation as described above, the diffusion equation 4.18 is solved with a uniform initial concentration and a zero boundary concentration.

#### 4.4.1 Analytical Solutions ( $m_1 = 0$ ).

The analytical solutions for massive and hollow systems with zero boundary concentration can be found in literature [37-39] and are summarized in Appendix B. In this main text only the solutions for massive systems are presented.

##### Slab ( $\nu=0$ )

$$E' = 1 - 2 \sum_1^{\infty} \frac{\cos(\mu_k y)}{\sin(\mu_k)} \frac{\exp(-\mu_k^2 \tau)}{\mu_k} \quad (4.54)$$

in which  $\mu_k$  are the positive roots of the characteristic equation  $\cos(\mu_k) = 0$ ; thus  $\mu_k = (k - \frac{1}{2})\pi$  and  $\sin(\mu_k) = (-1)^{k+1}$ .

##### Massive Cylinder ( $\nu=1$ )

$$E' = 1 - 2 \sum_1^{\infty} \frac{J_0(\mu_k y)}{J_1(\mu_k)} \frac{\exp(-\mu_k^2 \tau)}{\mu_k} \quad (4.55)$$

in which  $J_0$  and  $J_1$  are Bessel functions [41] of the first kind of

order 0 respectively order 1;  $\mu_k$  are the positive roots of  $J_0(\mu_k) = 0$ .

A quite well approximation of the first six roots is given by

$\mu_k = 2.4048 + (k-1)\pi$  with deviations  $\leq 0.5\%$ .

#### Massive Sphere ( $\nu=2$ )

$$E' = 1 + \frac{2}{y} \sum_1^{\infty} \frac{\sin(\mu_k y)}{\cos(\mu_k)} \frac{\exp(-\mu_k^2 \tau)}{\mu_k} \quad (4.56)$$

in which  $\mu_k$  are the positive roots of  $\sin(\mu_k) = 0$ ; thus  $\mu_k = k\pi$  and  $\cos(\mu_k) = (-1)^k$ .

From the above solutions the following expressions for the flux and the efficiency can be derived :

$$F = -\left(\frac{\partial m}{\partial y}\right)_{y=1} = \left(\frac{\partial E'}{\partial y}\right)_{y=1} = 2 \sum_1^{\infty} \exp(-\mu_k^2 \tau) \quad (4.57)$$

and

$$1-E = \int_{\tau}^{\infty} F X_{i,\sigma=0} d\tau = 2(\nu+1) \sum_1^{\infty} \frac{\exp(-\mu_k^2 \tau)}{\mu_k^2} \quad (4.58)$$

#### 4.4.2 Penetration Period ( $m_1 = 0$ ).

At small  $\tau$ -values concentration profiles have not yet penetrated into the centre of the material and a slab may be looked upon as an infinite body. Moreover, cylinders and spheres behave like a slab in case of extremely small  $\tau$ -values ( $\tau \rightarrow 0$ ). For these situations quite a number of terms of the series solutions (eqns. 4.54-4.56) is required. A simple short-time solution can be obtained from the analytical

solution of the diffusion equation for infinite slabs ( $-\infty < y < 1$ ) with a homogeneous initial concentration and a zero boundary concentration [37]:

$$E' = 1 - \operatorname{erf}\left(\frac{1-y}{2\sqrt{\tau}}\right) \quad (4.59)$$

For the flux follows:

$$F = \left(\frac{\partial E'}{\partial y}\right)_{y=1} = \frac{1}{\sqrt{(\pi\tau)}} \quad (4.60)$$

and for the drying efficiency:

$$E = \int_0^{\tau} F X_{i,\sigma=0} d\tau = 2X_{i,\sigma=0} \frac{\sqrt{\tau}}{\sqrt{\pi}} \quad (4.61)$$

Elimination of  $\tau$  from equations 4.60 and 4.61 gives the relation wanted between  $F$  and  $E$ :

$$F = \frac{2}{\pi} X_{i,\sigma=0} \frac{1}{E} \quad (4.62)$$

This equation is valid during the entire Penetration Period of slabs, but for cylinders and spheres, as already mentioned, only for extreme small  $\tau$ -values ( $\tau \rightarrow 0$ ).

Applying the definition of the  $G$ -parameter (eqn. 4.35) now gives (realise that  $m_i = 0$ , thus  $E'_i = 1$ ):

$$G_0 = \frac{2}{\pi} \quad (4.63)$$

and it appears that also in this situation  $G_0$  is independent of geometry and hollowness.

The entire Penetration Period for all hollow and massive geometries can be described very well with the correlation:

$$F = G_0 X_{i, \sigma=0} \left[ \frac{1}{E} - \beta \right] \quad (4.64)$$

in which for the correlation parameter  $\beta$  the following relations can be obtained by fitting:

$$\text{For a slab} \quad : \quad \beta = 0 \quad (4.65a)$$

$$\text{For a cylinder:} \quad \beta = 0.71 - 0.13\lambda - 0.58\lambda^2 \quad (4.65b)$$

$$\text{For a sphere} \quad : \quad \beta = 0.88 - 0.08\lambda - 0.08\lambda^2 - 0.72\lambda^3 \quad (4.65c)$$

In terms of the G-parameter equation 4.64 reads:

$$G = G_0(1 - \beta E) \quad (4.66)$$

It can be concluded now, that penetration processes with both constant boundary flux and constant boundary concentration can be described in a similar way.

#### 4.4.3 Regular Regime ( $m_1 = 0$ ).

At sufficiently high  $\tau$ -values the analytical solutions of §4.4.1 can be approximated quite well by the first term of the series. The concentration profiles take on the shape of a cosine, Bessel or damped sine function and decrease exponential with time. This shape no longer changes (similarity of concentration profiles) and the drying behaviour has become independent of the initial conditions; sooner or later, every drying process will arrive at this drying stage, which is called: "Regular Regime with constant boundary concentration".

The Regular Regime solution for the flux reads:

$$F = 2\exp(-\mu_1^2 \tau) \quad (4.67)$$

and for the efficiency:

$$1-E = 2(\nu+1) \frac{\exp(-\mu_1^2 \tau)}{\mu_1^2} \quad (4.68)$$

Elimination of  $\tau$  from those two equations gives a simple expression for the relation between  $F$  and  $E$  :

$$F = \frac{\mu_1^2}{\nu+1} (1-E) \quad (4.69)$$

Analog to equation 4.47 the flux versus efficiency can also be expressed as (remember  $m_1=0$ , so  $E'_1=1$ ):

$$F = \frac{1}{2} \text{Sh}_d (1-E) \quad (4.70)$$

From equations 4.69 and 4.70 it follows for massive geometries:

$$\text{Sh}_d = \frac{2\mu_1^2}{\nu+1} \quad (4.71)$$

In a similar way, for both massive and hollow geometries, one finds:

$$\text{Sh}_d = \frac{2\mu_1^2}{\bar{X}_{i,\sigma=0}} \quad (4.72)$$

in which  $\mu_1$  follows from the characteristic equations as given in the

## Appendix C.

Apparently, during the Regular Regime with constant boundary concentration  $Sh_d$  takes on a constant value. Furthermore, it can be concluded that the Regular Regimes with constant boundary flux and constant boundary concentration can be described in a similar way.

In Table 4.3 values of  $Sh_d$  are summarized. As expected, the values for a hollow geometry lay between those of the massive geometry and the slab. Note that at  $\lambda=0.6-0.7$  a minimum value of  $Sh_d$  occurs! The exact  $Sh_d$  values of Table 4.3 can be correlated quite well by the following polynoms of  $\lambda$ :

$$\text{For a slab} \quad : \quad Sh_d = \frac{\pi^2}{2} = 4.935 \quad (4.73a)$$

$$\text{For a cylinder} \quad : \quad Sh_d = 5.75 - 4.36\lambda + 6.34\lambda^2 - 2.82\lambda^3 \quad (4.73b)$$

$$\text{For a sphere} \quad : \quad Sh_d = 6.58 - 7.14\lambda + 8.78\lambda^2 - 3.29\lambda^3 \quad (4.73c)$$

The maximum deviations between the exact and approximated values of  $Sh_d$  amount 0.7% for cylinders and 0.4% for spheres.

**Table 4.3:** Values of  $Sh_d$  during Regular Regime with constant boundary concentration ( $D_r=1, \sigma=0, m_i=0$ ).

$\lambda$	cylinder ( $\nu=1$ )		sphere ( $\nu=2$ )	
	exact (eqn. 4.72)	approx. (eqn. 4.73)	exact (eqn. 4.72)	approx. (eqn. 4.73)
0	5.783	5.750	6.580	6.580
0.1	5.339	5.375	5.953	5.951
0.2	5.087	5.109	5.462	5.477
0.3	4.943	4.936	5.127	5.139
0.4	4.864	4.840	4.918	4.918
0.5	4.828	4.803	4.802	4.794
0.6	4.819	4.807	4.752	4.746
0.7	4.830	4.837	4.752	4.756
0.8	4.856	4.876	4.787	4.803
0.9	4.891	4.906	4.850	4.867
1	4.935	4.910	4.935	4.930

massive
slab ( $\nu=0$ )



#### 4.4.4 Transition from Penetration Period to Regular Regime ( $m_1 = 0$ ).

Again it appears that both extremes can be connected most simply by just stepping over from the approximation of the one extreme to that of the other. Further, it has practical advantages now to formulate the transition criterium in terms of efficiencies, in other words: the Penetration Period ends and the Regular Regime starts at some specified value of the efficiency ( $E=E_T$ ). From the exact calculated values of F versus E the following transition criterium is found:

$$E_T = 0.5 + 0.05\nu(5-\nu)(1-\lambda) \quad (4.74)$$

#### 4.4.5 Algorithm for Short-Cut Calculation ( $m_1 = 0$ ).

Expressions for the drying time can now be found from equations 4.23, 4.64 and 4.70:

##### Penetration Period ( $E < E_T$ ):

$$\text{For a slab} \quad : \tau = \frac{E^2}{2G_0} \quad (4.75)$$

$$\text{For a cylinder and a sphere} \quad : \tau = \frac{-\beta E - \ln(1-\beta E)}{G_0 (X_{i,\sigma=0})^2 \beta^2} \quad (4.76)$$

##### Regular Regime ( $E > E_T$ ):

$$\text{For all geometries} \quad : \tau = \tau_T + \frac{2}{\text{Sh}_d X_{i,\sigma=0}} \ln \left[ \frac{1-E_T}{1-E} \right] \quad (4.77)$$

where  $\tau_T$  is the drying time at the end of the Penetration Period, when transition takes place.

Assuming that the drying times of stage I and II are negligible with respect to the total drying time, the following algorithm could be used for the calculation of the drying flux  $F$  and the drying time  $\tau$  at any given value of the drying efficiency  $E$ .

```

begin {short-cut algorithm,  $D_r=1$ ,  $\sigma=0$ ,  $m_1=0$ , isothermal}
  read ( $v, \lambda, E$ )
  calculate  $\beta$  from equations 4.65a-4.65c
  calculate  $E_T$  from equation 4.74
  if  $E \leq E_T$ 
    then {Penetration Period}
      calculate  $F$  from equation 4.64
      calculate  $\tau$  from equations 4.75 or 4.76
    else {Regular Regime}
      calculate  $\tau_T$  from equations 4.75 or 4.76
      calculate  $Sh_d$  from equations 4.73
      calculate  $F$  from equations 4.70
      calculate  $\tau$  from equations 4.77
  write ( $F, \tau$ )
end.
```

Comparison of this short-cut calculation with the exact calculation shows that maximum errors in the flux and in the drying time occur at the transition point. It should be noticed that at the transition point the flux calculated with the penetration correlation yields a too high value, whereas the regular regime correlation gives a too low value. In any case, the relative deviation between both approximate fluxes and the the exact flux is within 5% .

*The maximum errors in the approximated drying times are only 3% .*

#### 4.5 Adiabatic Drying Process.

During the constant activity period of an adiabatic drying process the drying system takes on the *wet-bulb-temperature*  $T_{wb}$ . After this *isothermal* period the temperature of the system will increase. As was pointed out in literature [23,26,33] heat transfer from a gas phase to a body is very slow with respect to heat diffusivity in the body. This means that the temperature distribution in the body will remain nearly uniform, so the averaged temperature  $\bar{\theta}$  of the body nearly equals the surface temperatures  $\theta_i$ ; and because equilibrium is assumed at the interface:  $\theta \approx \bar{\theta} \approx \theta_i = \theta'_i$ , where  $\theta'_i$  is the interface temperature in the gas phase.

Because in general the (latent) heat of evaporation ( $L$ ) of the moisture is many times larger than the heat capacity of the body, a negligible part of the total heat flux is left to accumulate in the body. From this it follows that by good approximation the heat transfer process may be considered as a *quasi steady-state adiabatic evaporation process*, for which the following thermal energy balance holds:

$$\alpha(\theta'_\infty - \theta'_i) \approx j_{mi}^s L_i \quad (4.78)$$

Note that this equation represents the wet bulb equation in its elementary form.

In case of an enclosed gas phase, hollow systems will blow up with increasing temperature; however, this complication will not be considered here and the internal and external radii ( $R_1$  and  $R_2$ ) are assumed to remain constant.

The diffusion coefficient  $D_0$  depends on the temperature and so changes during the drying process. The mass balance over the drying system now reads:

$$FX_{i,\sigma=0} \frac{D_0}{(R_2-R_1)^2} dt = dE \quad (4.79)$$

and the dimensional drying time  $t$ :

$$t = \frac{(R_2-R_1)^2}{X_{i,\sigma=0}} \int_0^E \frac{dE}{FD_0} \quad (4.80)$$

Assuming a constant value of the hollowness factor  $\lambda$ , which is certainly true for massive bodies, the relationship  $F$  versus  $E$  (§4.4) is independent of temperature. In order to keep a maximum attainable value 1 for the efficiency  $E$ , the equilibrium moisture content  $\rho_{m^*}$  in the definitions of the fluxparameter  $F$  and the efficiency  $E$  should be taken from the sorption isotherm at dry bulb temperature. The temperature dependence of the diffusion coefficient  $D_0$  can often be expressed by an Arrhenius type relation:

$$D_0(T) = D_0(T_1) \exp\left[-\frac{A_D}{R}\left(\frac{1}{T} - \frac{1}{T_1}\right)\right] \quad (4.81)$$

in which  $T$  is the absolute temperature of the body ( $^{\circ}\text{K}$ ),

$A_D$  is the activation energy for diffusion [Joule/mol $^{\circ}\text{K}$ ] and

$R$  is the gas constant.

From the definition of the flux parameter (eqn.4.11) and the thermal energy balance (eqn.4.78) the following relation for  $FD_0$  can be derived:

$$\frac{FD_0}{(FD_0)_{ca}} = \frac{j_{mi}^s}{(j_{mi}^s)_{ca}} = \frac{\alpha}{\alpha_{ca}} \frac{L_{ca}}{L_i} \frac{\theta'_{\infty} - \theta}{\theta'_{\infty} - \theta_{ca}} \quad (4.82)$$

in which the subscript "ca" indicates the value of the parameters at

the condition of constant surface activity (thus  $\theta_{ca}$  is the wet bulb temperature); parameters without this subscript have actual values. If the difference between the dry- and wet-bulb temperature is not too high, the heat transfer coefficient  $\alpha$  and the evaporation enthalpy  $L$  will not change too much and equation 4.82 simplifies to:

$$\frac{FD_0}{(FD_0)_{ca}} \approx \frac{\theta'_\infty - \theta}{\theta'_\infty - \theta_{ca}} = \frac{T'_\infty - T}{T'_\infty - T_{ca}} \quad (4.83)$$

Assuming again very short stages I and II the drying curves  $F$  versus  $E$ , as given by equations 4.64 and 4.70, have to be used and the drying time can be calculated from equation 4.80; however, now the integration has to be carried out numerically (e.g. trapezium rule).

The relationship  $FD_0$  versus  $E$  can be found by the following procedure:

- choose a value for  $T$  (remember:  $T_{ca} \leq T \leq T'_\infty$ )
- calculate  $FD_0$  from equation 4.82 or 4.83
- calculate  $D_0$  from equation 4.81
- calculate  $F$  from the values of  $FD_0$  and  $D_0$
- calculate  $E$  from the temperature independent relationship  $F$  versus  $E$  (eqns. 4.64 and 4.70)

The above procedure yields at any given temperature  $T$  the values of  $E$ ,  $F$  and  $D$ . If one desires to integrate with equal steps of  $E$ , the temperature  $T$  must be found by iteration from equation 4.83.



## CHAPTER V

### POWER LAW DIFFUSION IN SYSTEMS WITH ANY DEGREE OF SHRINKAGE

#### 5.1 Concentration Dependence of Diffusion Coefficient.

It is assumed that the moisture diffusion coefficient depends on the moisture concentration according to a power law relation (see also chapter I):

$$D\rho_s^2 = b \left[ \frac{\rho_m}{\rho_s} - \frac{\rho_{m\#}}{\rho_{s\#}} \right]^a = b(u-u_{\#})^a \quad (5.1)$$

$\rho_{m\#}$  is the moisture concentration where the diffusion coefficient becomes practically zero; this thesis only deals with systems in which the diffusion coefficient approaches to zero at the equilibrium moisture content, so that  $u_{\#} \approx u_{*}$ .

$a$  and  $b$  are fitting parameters, which can be found by linear regression of  $\ln(D\rho_s^2)$  versus  $\ln(u-u_{\#})$ . In chapter VI it will be pointed out how these fitting parameters can be derived from a drying experiment.

The diffusion coefficient  $D_0$  at the initial concentration  $u_0$  is given by:

$$D_0\rho_{s0}^2 = b(u_0-u_{\#})^a \quad (5.2)$$

Applying the definitions of the reduced diffusion coefficient  $D_r$  and the concentration  $m$  (§3.3) yields:

$$D_r = m^a \quad (5.3)$$

It will be clear that for *non-shrinking* systems, a concentration independent diffusion coefficient ( $D_r=1$ , so  $a=0$ ) is a special case of power law diffusion. However, it should be noticed that for *shrinking* systems  $a=0$  does *not* correspond with a constant diffusion coefficient.

## 5.2 Numerical Solution of the Diffusion Equation.

The power relation for  $D_r$  has to be substituted in the generalized diffusion equation with  $\phi$ -coordinates (§3.3) or in the diffusion equation with linear coordinates (§4.1).

In general, the diffusion equation, both in  $\phi$ -coordinates and linear coordinates, requires a numerical approach to be solved. It is beyond the scope of this thesis to deal extensively with the numerical approach. Only some important aspects, which may cause serious problems, are treated in Appendix D.

In general the solution of the diffusion equation (§3.3) can be written as:

$$m = m(\phi, \tau, v, \lambda, a, \sigma_{v_0}, \sigma_{v_{\#}}, \text{drying stage}) \quad (5.4)$$

and the averaged concentration is given by:

$$\bar{m} = \bar{m}(\tau, v, \lambda, a, \sigma_{v_0}, \sigma_{v_{\#}}, \text{drying stage}) \quad (5.5)$$



The drying time is calculated by integration of the mass balance (§3.4); the required relationship  $F$  versus  $E$  is now obtained from the numerical computer output and will be a function of the following form:

$$F = F(E, \nu, \lambda, a, \sigma v_0, \sigma v_{\#}, \text{drying stage}) \quad (5.6)$$

For didactical reasons the drying stage with constant surface concentration will be discussed first.

### 5.3 Drying Stage with Constant Surface Concentration ( $m_i = 0$ ).

For this drying stage Liou [28,29] proposed short-cut correlations for  $F$  versus  $E$ , which are valid for slabs, massive cylinders and massive spheres. The Regular Regime is described with Sherwood numbers, whereas the Penetration Period of non-shrinking systems is described with Taylor series expansions for the  $G$ -parameter. In this section the method, used by Liou, is simplified and extended to hollow bodies, irrespective of their degree of shrinkage.

#### 5.3.1 Non-Shrinking Systems ( $D_r = m^a, \sigma = 0, m_i = 0$ ).

##### Penetration Period

It appears that during this period  $G$  versus  $E$  can be approximated quite well by a linear relationship. During the Penetration Period the expressions for the drying flux and drying time are similar to those of constant diffusion coefficient, however, the parameters  $G_0$  and  $\beta$  now depend on the exponent  $a$ :

$$F = G_0 X_{i, \sigma=0} \left( \frac{1}{E} - \beta \right) \quad (5.7)$$

For a slab: 
$$\tau = \frac{E^2}{2G_0} \quad (5.8)$$

For a cylinder:  
and a sphere 
$$\tau = \frac{-\beta E - \ln(1-\beta E)}{G_0(X_{i,\sigma=0})^2 \beta^2} \quad (5.9)$$

in which:

$$G_0 = \frac{2}{\pi} \left[ \frac{1.42}{a+1.42} \right]^{1.98} \quad \text{for } 0 \leq a < \infty \quad (5.10)$$

and

$$\beta = \beta_{a=0} (1.25)^a \quad \text{for } 0 \leq a < 2 \quad (5.11)$$

The expressions for  $\beta_{a=0}$  are given by equations 4.65.

Regular Regime ( $D_r = m^a$ ,  $\sigma = 0$ ,  $m_i = 0$ )

During this drying period mass transfer is best described by means of a Sherwood number according to equations 4.45 and 4.47. For power law diffusion and a zero boundary concentration the flux equation now becomes:

$$F = \frac{1}{2} \frac{Sh_d}{a+1} (1-E)^{a+1} \quad (5.12)$$

Schoeber [23] observed that in case of power law diffusion ( $D_r = m^a$ ) during the Regular Regime the Sherwood numbers ( $Sh_d$ ) have constant values. Liou [28,29] reports that those constant values depend pseudo linearly on  $\frac{a}{a+2}$ . (However, both Schoeber and Liou did not recognize that constant values of  $Sh_d$  will not occur in case of a non zero boundary concentration!)

Both observations are quite important, because they enable the development of very accurate correlations for the Regular Regime, being the drying stage that contributes predominantly to the drying time. A similar behaviour may be expected for hollow systems. Indeed, from the numerical computer output the following correlations for  $Sh_d$  can be derived:

$$Sh_d = Sh_{d,a \rightarrow \infty} - \Delta Sh_d \cdot \frac{2}{a+2} \quad (5.13)$$

in which:

$$\Delta Sh_d = Sh_{d,a \rightarrow \infty} - Sh_{d,a=0} \quad (5.14)$$

$$Sh_{d,a \rightarrow \infty} = 7.391 + (3.516v - 0.034) \left[ \frac{X_{i,\sigma=0} - 1}{v} \right]^{1.535 - 0.075v} \quad (5.15)$$

$Sh_{d,a=0}$  is given by  $\lambda$ -polynoms according to equations 4.73. However, a more elegant correlation for  $\Delta Sh_d$ , based on normalized  $X_i$ -values, reads:

$$\Delta Sh_d = 2.456 + (2.720v - 0.087) \left[ \frac{X_{i,\sigma=0} - 1}{v} \right]^{1.04} \quad (5.16)$$

Note: only the exponents in equations 5.15 and 5.16 follow from a regression analysis.

*The accuracy of the above correlation is within  $\pm 1\%$ .*

The Regular Regime expressions for the drying time (isothermal) follow from the integration of the mass balance with the aid of equation 5.12:

$$\text{For } a=0 \quad : \quad \tau = \tau_T + \frac{2}{\text{Sh}_d X_{i,\sigma=0}} \ln \left[ \frac{1-E_T}{1-E} \right] \quad (5.17)$$

(eqn. 4.77)

$$\text{For } a \neq 0: \quad \tau = \tau_T + \frac{2}{\text{Sh}_d X_{i,\sigma=0}} \frac{a+1}{a} \left[ \frac{1}{(1-E)^a} - \frac{1}{(1-E_T)^a} \right] \quad (5.18)$$

Transition ( $D_r = m^a$ ,  $\sigma = 0$ ,  $m_i = 0$ )

The Liou approach needs a quite severe criterium to be sure of Regular Regime behaviour, namely:

$$E \geq \frac{2}{a+2} \quad (5.19)$$

So, in case of a concentration independent diffusion coefficient ( $a=0$ ) the criterium becomes  $E=1$ , and the whole drying process is considered as a penetration process. However, the more simple approach described in this thesis takes better advantage of the Regular Regime properties, as given in Chapter IV.

From the numerical calculated solutions the following transition criterium has been derived:

$$E_T = \frac{1+0.1v(5-v)(1-\lambda)}{a+2} \quad (5.20)$$

### 5.3.2 Systems with any Degree of Shrinkage ( $D_r = m^a$ , $0 \leq \sigma \leq 1$ , $m_i = 0$ )

At the same value of the drying efficiency  $E$ , the flux parameter  $F$  of the shrinking system ( $\sigma > 0$ ) is related to the flux parameter  $F_{\sigma=0}$  of the non-shrinking system by means of the shrinkage factor  $H$ , introduced by Liou [28,29]:

$$H = \frac{F}{F_{\sigma=0}} \quad (5.21)$$

Finding  $F$  versus  $E$  has been reduced now to finding expressions for the shrinkage factor  $H$ .

For a slab geometry ( $\nu=0$ ) the solutions of the generalized diffusion equation (53.4) are independent of the shrinkage behaviour, because in all cases  $X_{\phi} = X_1 = X_{1,\sigma=0} = 1$ . So the solutions of non-shrinking slabs, expressed in dimensionless quantities, are identical with those of shrinking slabs. The shrinkage properties of the material now emerge from the definitions of the dimensionless parameters and the diffusion relation (eqn. 5.1).

*Conclusion: the shrinkage factor of slabs is 1, irrespective of the drying stage.*

For cylinders and spheres correlations of the shrinkage factor were derived according to Liou's method: first find the initial value ( $\tau \rightarrow 0$ ), then look for an approximate solution for large times and eventually connect both extreme situations by means of a Taylor series expansion.

Penetration with extremely short times ( $D_r = m^a$ ,  $0 \leq \sigma \leq 1$ ,  $m_1 = 0$ )

The flux parameter of the shrinking system (eqn. 3.14) is related to the flux parameter of a (fictive) non-shrinking system (eqn. 4.11) with exactly the same initial conditions and the same product properties. From the definitions of the flux parameters (eqns. 3.14 and 4.11) follows:

$$\lim_{\tau \rightarrow 0} H = H_0 = \frac{d_{s,ap} R_s}{\rho_{s0} (R_{2,0} - R_1)} \quad (5.22)$$

Equation 5.22 can also be written as:

$$H_0 = \frac{A_0 R_s}{V_s} \cdot \frac{d_{s,ap} V_s}{\rho_{s0} V_0} \cdot \frac{V_0}{A_0 (R_{2,0} - R_1)} \quad (5.23)$$

where  $A_0$  and  $V_0$  are the initial values of the surface area and the (shell-)volume of the system.

From the solid mass balance  $d_{s,ap} V_s = \rho_{s0} V_0$  and from the definitions of  $X_i$  (eqns. 3.40 and 4.16) follows:

$$H_0 = \frac{X_{i,0}}{X_{i,\sigma=0}} \quad (5.24)$$

Substituting the expression for  $X_{i,0}$  (eqn. 3.43) gives:

$$H_0 = \left[ 1 + (1 - \lambda^{\nu+1}) \sigma V_0 \right]^{\frac{\nu}{\nu+1}} \quad (5.25)$$

This expression confirms that for a slab ( $\nu=0$  or  $\lambda \rightarrow 1$ ) the shrinkage factor  $H_0=1$ .

**Note:**

Applying the definition of the G-parameter (eqn. 4.35) to a shrinking system with zero boundary concentration yields:

$$G_0 = \lim_{\tau \rightarrow 0} \frac{FE}{X_i} = \lim_{\tau \rightarrow 0} \frac{HF_{\sigma=0} E}{X_i} = \lim_{\tau \rightarrow 0} \frac{H_0 F_{\sigma=0} E}{X_{i,0}} \quad (5.26)$$

with the aid of equation 5.24:

$$G_0 = \lim_{\tau \rightarrow 0} \frac{F_{\sigma=0} E}{X_{i,\sigma=0}} \quad (5.27)$$

Conclusion: the  $G_0$ -correlation for non-shrinking systems (eqn. 5.10) is valid for shrinking systems as well.

Regular Regime ( $D_r = m^a$ ,  $0 \leq \sigma \leq 1$ ,  $m_1 = 0$ )

A relation for the initial shrinkage factor  $H_0$  could be found by an algebraic analysis. However, the Regular Regime correlations for the shrinkage factor has to be deduced from the numerical computer output. The Sherwood numbers for shrinking systems, which do not take on constant values during the Regular Regime, are described by Schoeber [23] in the following way:

$$Sh_d = Sh_{d,\sigma=0} + \Delta Sh_d \quad (5.28)$$

and the shrinkage factor simply follows:

$$H = 1 + \frac{\Delta Sh_d}{Sh_{d,\sigma=0}} \quad (5.29)$$

For strongly concentration dependent diffusion coefficients ( $a \rightarrow \infty$ ) the concentration profile is rectangular ( $m = \bar{m}$ ) and  $\Delta Sh_{d,a \rightarrow \infty}$  can be calculated analytically by comparing the flux expressions:

$$F = -D_r X_i \left( \frac{\partial m}{\partial \Phi} \right)_{\Phi=1} \quad (5.30)$$

$$F_{\sigma=0} = -D_r X_{i,\sigma=0} \left( \frac{\partial m}{\partial \Phi} \right)_{\Phi=1} \quad (5.31)$$

from which follows:

$$H_{a \rightarrow \infty} = \frac{X_i}{X_{i,\sigma=0}} \quad (5.32)$$

and from equations 5.29 and 5.32 with  $Sh_{d,\sigma=0} = Sh_{d,a \rightarrow \infty}$ :

$$\Delta Sh_{d,a \rightarrow \infty} = Sh_{d,a \rightarrow \infty} \left[ \frac{X_i}{\bar{X}_{i,\sigma=0}} - 1 \right] \quad (5.33)$$

Schoeber [23] observed that  $\Delta Sh_d$  is hardly influenced by the kind of concentration dependence of the diffusion coefficient, so

$\Delta Sh_d \approx \Delta Sh_{d,a \rightarrow \infty}$  and from equations 5.29 and 5.33 now follows with

$$Sh_{d,\sigma=0} = Sh_{d,a}$$

$$H = 1 + \frac{Sh_{d,a \rightarrow \infty}}{Sh_{d,a}} \left[ \frac{X_i}{\bar{X}_{i,\sigma=0}} - 1 \right] \quad (5.34)$$

Substitution of the expressions for  $X_i$  (eqn. 3.42) and  $H_0$  (eqn. 5.25) finally yields for the shrinkage factor:

$$H = 1 + \frac{Sh_{d,a \rightarrow \infty}}{Sh_{d,a}} \left[ H_0 (1-sE)^{\frac{v}{v+1}} - 1 \right] \quad (5.35)$$

The Sherwood numbers in this equation are to be calculated with equations 5.13-5.16.

From equation 5.35 it can be seen that the shrinkage factor  $H$  decreases as the drying efficiency  $E$  increases. If the averaged moisture content  $\bar{v} = v_x = 0$ , the shrinkage factor reaches the minimum value 1. Thus, at decreasing moisture concentrations the shrinking abilities of the material become less and at very low moisture concentrations the shrinking system even behaves like a non-shrinking system.



Transition Period ( $D_r = m^a$ ,  $0 \leq \sigma \leq 1$ ,  $m_i = 0$ )

According to the criterium, used in Liou's approach [28,29], the Regular Regime correlation 5.35 may be used if  $E \geq E_Q$ , with:

$$E_Q = \frac{a}{a+2} \quad (5.36)$$

For the transition region ( $0 < E < E_Q$ ) the shrinkage factor  $H$  is approximated by an appropriate Taylor series expansion, starting in ( $E=0$ ,  $H=H_0$ ) and merging into the Regular Regime correlation at  $E=E_Q$ . To obtain a smooth transition at  $E=E_Q$ , equal zero and first derivatives of the Regular Regime correlation and the Taylor function at  $E=E_Q$  are imposed:

$H=H_Q$  (equal zero derivatives) and

$H^{(1)}=H_Q^{(1)}$  (equal first derivatives).

The following Taylor series expansion appears to be a satisfying approximation of the transition region:

$$H = H_0 + \frac{H_0^{(2)}}{2} E^2 + \frac{H_0^{(3)}}{6} E^3 \quad (5.37)$$

where  $H_0^{(2)}$  and  $H_0^{(3)}$  result from the above two conditions at  $E=E_Q$ :

$$H_0^{(2)} = \frac{6}{E_Q^2} \left[ H_Q - H_0 - \frac{E_Q}{3} H_Q^{(1)} \right] \quad (5.38)$$

$$H_0^{(3)} = \frac{2}{E_Q^2} \left[ H_Q^{(1)} - E_Q H_0^{(2)} \right] \quad (5.39)$$

Drying times ( $D_r = m^a$ ,  $0 \leq \sigma \leq 1$ ,  $m_i = 0$ )

The drying time follows from the integration of the mass balance:

$$\tau = \int_0^E \frac{dE}{FX_i} = \int_0^E \frac{dE}{HF_{\sigma=0} X_i} \quad (5.40)$$

where,  $F_{\sigma=0}$  is given by equation 5.7 for  $0 \leq E \leq E_T$  and  
by equation 5.12 for  $E > E_T$

$H$  is given by equations 5.37 for  $0 \leq E \leq E_Q$  and  
by equation 5.35 for  $E > E_Q$

$X_i$  is given by equation 3.42

In general this integration has to be carried out by means of numerical methods (e.g. trapezium rule).

#### 5.4 Drying Stage with Constant Surface Activity.

##### 5.4.1 Non-Shrinking Systems ( $D_r = m^a$ , $\sigma = 0$ , $F_{ca} \approx \text{constant}$ )

During the period with constant surface moisture activity ( $a_{mi} \approx \text{constant}$ ) the drying flux of a non-shrinking system will remain nearly constant:  $F = F_{ca} \approx \text{constant}$  (see chapter IV).

The contents of §4.3 also apply to systems with concentration dependent diffusion coefficients. However, the relationship among  $F_{ca}$ ,  $E$  and  $E'_i$  has to be derived now from the numerical computer output.

A straight forward use of the G-parameter (eqn. 4.35) and of correlations according to equation 4.37 now fails, because the parameter  $E/E'_i$  no longer is a continuously increasing quantity in all drying situations, e.g. at high fluxes  $E/E'_i$  versus  $\tau$  may show a maximum!

A different but successful approach for power law diffusion departs

from a transformation of the generalized diffusion equation with  
 $\tilde{m} = m^{a+1}$  (see also Appendix D):

Generalized partial differential equation:

$$\frac{\partial \tilde{m}}{\partial \tau} = (\tilde{m})^{\frac{a}{a+1}} \frac{\partial}{\partial \Phi} (X^2 \frac{\partial \tilde{m}}{\partial \Phi}) \quad (5.41)$$

Initial condition:

$$\tau = 0 \quad 0 \leq \Phi < 1 \quad \tilde{m} = 1 \quad (5.42)$$

Boundary conditions:

$$\tau > 0 \quad \Phi = 0 \quad X \frac{\partial \tilde{m}}{\partial \Phi} = 0 \quad (5.43)$$

$$\Phi = 1 \quad -X_i \frac{\partial \tilde{m}}{\partial \Phi} = \tilde{F} \quad (5.44)$$

where  $\tilde{F} = F(a+1)$ .

The solution of this transformed diffusion equation can be written as:

$$\tilde{m} = \tilde{m}(\Phi, \tau, \nu, \lambda, \frac{a+1}{a}, \tilde{F}, \sigma_{v_0}, \sigma_{v_{\#}}) \quad (5.45)$$

For high  $a$ -values a limit solution is approached, because  $\frac{a}{a+1} \rightarrow 1$ .

In addition to  $\tilde{m}$  and  $\tilde{F}$  some more helpful parameters are defined below.

**Table 5.1** Definitions of some parameters.

$\tilde{m} = m^{a+1}$	$\tilde{F} = F(a+1)$
$\tilde{E} = 1 - (\tilde{m})^{a+1}$	$\tilde{E}_i = 1 - (m_i)^{a+1}$
$\epsilon = \frac{\tilde{E}}{\tilde{E}_i} = \frac{1 - (\tilde{m})^{a+1}}{1 - (m_i)^{a+1}}$	$\tilde{G} = \frac{\tilde{F} \tilde{E}}{\tilde{E}_i X_i}$

Regular Regime ( $D_r = m^a$ ,  $\sigma = 0$ ,  $F_{ca} \approx \text{constant}$ )

From equations 4.45 and 4.46 now follows:

$$F_{ca} = \frac{1}{2} \frac{Sh_{d,ca}}{a+1} \left[ (\bar{m})^{a+1} - (m_1)^{a+1} \right] \quad (5.46)$$

and expressed in the parameters of Table 5.1:

$$\tilde{F}_{ca} = \frac{1}{2} Sh_{d,ca} (\tilde{E}'_1 - \tilde{E}) \quad (5.47)$$

This latter expression for the flux parameter is analogous to equation 4.47 for concentration independent diffusion coefficient. However, in case of power law diffusion with  $a \neq 0$  no constant value for the Sherwood number is obtained: after passing through a minimum value (Figure 5.1) the Sherwood number is gradually increasing until an end value at  $E'_1=1$  is reached. Schoeber [23] observed that this end value is nearly independent of the flux parameter  $F_{ca}$ . For massive geometries Liou [28,29] found a pseudo linear relationship by plotting the numerically computed values of  $Sh_{d,ca}$  at  $E'_1=1$  against values of  $\frac{a}{a+2}$ ; it appears now that this linearity also holds for hollow geometries. The correlation for  $Sh_{d,E'_1=1}$  then reads:

$$Sh_{d,E'_1=1} = Sh_{d,a=0} + (Sh_{d,a \rightarrow \infty} - Sh_{d,a=0}) \frac{a}{a+2} \quad (5.48)$$

in which  $Sh_{d,a=0}$  is given by equation 4.51:

$$Sh_{d,a=0} = 6 + 2v \left[ \frac{X_{1,\sigma=0} - 1}{v} \right]^{2.15 - 0.18v} \quad (4.51)$$

and

$$\text{Sh}_{d,a=\infty} = 10.443 + 5.935 \nu \left[ \frac{X_{i,\sigma=0} - 1}{\nu} \right]^{1.25 - 0.03\nu} \quad (5.49)$$

Note that also in equation 5.49 only the exponent is a correlation parameter.

For Regular Regime behaviour with  $E'_i \leq 1$  the Sherwood number of the constant activity period  $\text{Sh}_{d,ca}$  is approximated quite well with the correlation:

$$\text{Sh}_{d,ca} = \text{Sh}_{d,a=0} + \left[ \text{Sh}_{d,E'_i=1} - \text{Sh}_{d,a=0} \right] E'_i \quad (5.50)$$

From equations 5.50 and 5.48 finally follows:

$$\text{Sh}_{d,ca} = \text{Sh}_{d,a=0} + \left[ \text{Sh}_{d,a=\infty} - \text{Sh}_{d,a=0} \right] \frac{a}{a+2} E'_i \quad (5.51)$$

Penetration Period ( $D_r = m^a$ ,  $\sigma = 0$ ,  $F_{ca} \approx \text{constant}$ )

Approximations for this drying stage are based on Taylor series expansions of  $\tilde{G}$  versus  $\epsilon$  according to:

$$\tilde{G} = \tilde{G}_0 \left[ 1 + \tilde{\alpha}\epsilon + \tilde{\gamma}\epsilon^3 \right] \quad (5.52)$$

from which follows:

$$\frac{\tilde{F}_{ca}}{\tilde{E}'_i} = \tilde{G}_0 X_i \left[ \frac{1}{\tilde{\epsilon}} + \tilde{\alpha} + \tilde{\gamma}\epsilon^2 \right] \quad (5.53)$$

in which the initial value  $\tilde{G}_0$  appears to be independent of the power  $a$ . The solution can be derived analytically (Appendix E):

$$\tilde{G}_0 = \frac{\pi}{4} \quad (5.54)$$

From the numerical calculations the initial slope of  $\tilde{G}$  versus  $\epsilon$  proved to be pseudo linear with  $aF_{ca}$  and the following correlation for  $\tilde{\alpha}$  can be derived:

$$\tilde{\alpha} = \frac{0.5 aF_{ca} - \nu(1-\lambda)}{X_{i,\sigma=0}} \quad (5.55)$$

A correlation for  $\tilde{\gamma}$  is based on the end point ( $\epsilon=\epsilon_e, \tilde{G}=\tilde{G}_e$ ) of the Penetration Period:

$$\tilde{\gamma} = \frac{\tilde{G}_e/\tilde{G}_0 - 1 - \tilde{\alpha} \epsilon_e}{\epsilon_e^3} \quad (5.56)$$

With respect to the end of the Penetration Period two situations are to be considered:

1) the Penetration Period ends where the Regular Regime starts.

Similarly to a concentration independent diffusion coefficient the following transition criterium should be used:

$$\frac{\tilde{F}_{ca}}{\tilde{E}_i} = 1.5 \quad (5.57)$$

Elimination of  $\tilde{F}_{ca}$  from the Regular Regime approximation (eqn. 5.47) and the transition criterium (eqn. 5.57) gives:

$$\epsilon_e = 1 - \frac{3.0}{Sh_{d,ca}} \quad (5.58)$$

and from the definition of  $\tilde{G}$  (Table 5.1):

$$\tilde{G}_e = \frac{1.5}{X_{i,e}} \epsilon_e \quad (5.59)$$

Note, that in the approach of this chapter the transition criterium can be formulated independently of the geometry parameter  $\nu$  and the hollowness factor  $\lambda$  (compare with eqn. 4.52).

2) the Penetration Period ends if  $E'_i = E'_{i,cr}$

If  $\tilde{F}_{ca} \geq 1.5 \tilde{E}'_{i,cr}$  then the transition criterium (eqn. 5.57) indicates, that there will not be a Regular Regime (with a constant boundary flux) succeeding the Penetration Period. In this case the drying stage with a constant boundary flux ends with a concentration profile, which has not yet penetrated into the centre of the material. The end value  $\epsilon_e$  will be higher as  $\tilde{F}_{ca}$  is chosen higher. The critical point curve  $\tilde{F}_{ca}$  versus  $\epsilon_e$  with  $\tilde{E}'_{i,cr} = E'_{i,cr} = 1$  shows great similarity to the penetration curve (eqn. 5.7) of a drying process with a zero boundary concentration. However, a much better approximation is found if  $F_{ca}$  versus  $E_e$  is considered instead of  $\tilde{F}_{ca}$  versus  $\epsilon_e$ :

$$F_{ca} = G_{0,ca} X_i \left( \frac{1}{E_e} - \alpha \right) \quad (5.60)$$

For  $a=0$  this equation must be identical with the penetration curve of a drying proces with constant boundary flux and  $E'_i=1$  (eqn. 4.37)

From the numerical computer output the following correlations for

$G_{0,ca}$  and  $\alpha$  are derived:

$$G_{0,ca} = \frac{\pi}{4} \left[ \frac{1.45}{a+1.45} \right]^{1.89} \quad \text{for } 0 \leq a < \infty \quad (5.61)$$

$$\alpha = \alpha_{a=0} + \Delta\alpha \quad \text{for } 0 \leq a < 4 \quad (5.62)$$

where  $\alpha_{a=0}$  is given by equations 4.38; for  $a \neq 0$  a correction  $\Delta\alpha$ , which is pseudo linear with  $a$ , has to be added:

For a slab :  $\Delta\alpha = (0.11)a$  (5.63a)

For a cylinder:  $\Delta\alpha = (0.22-0.05\lambda-0.20\lambda^2+0.14\lambda^3)a$  (5.63b)

For a sphere :  $\Delta\alpha = (0.24+0.03\lambda-0.19\lambda^2+0.03\lambda^3)a$  (5.63c)

#### 5.4.2 Systems with Any Degree of Shrinkage

$$(D_r = m_-^a, 0 \leq \sigma \leq 1, a_{mi} \approx \text{constant}).$$

For shrinking cylinders and spheres the *drying flux will not remain constant during the period with constant surface water activity*, because the mass transfer coefficient in the gas phase depends on the dynamic outer radius  $R_{2,t}$  of the shrinking body. In general the following relation holds:

$$j_{wi,t}^s(R_{2,t})^q = \text{constant} = j_{wi,0}^s(R_{2,0})^q \quad (5.64)$$

where,

$q=0$  in case of a slab ( $v=0$ );

$q=1$  if the Sherwood number for the gasphase (Sh) is constant; for instance,  $Sh=2$  for a droplet in a spray drier.

$q=1-n$  if Sh depends on the Reynolds number according to  $Sh \sim R_e^n$  (eqn. 2.45)

The flux parameter of the constant activity period ( $F_{ca}$ ) can be related to its initial value ( $F_{ca,0}$ ) from equation 5.64 as follows:

$$\frac{F_{ca}}{F_{ca,0}} = \frac{j_{wi,t}^s}{j_{wi,0}^s} = \left[ \frac{R_{2,0}}{R_{2,t}} \right]^q = \left[ \frac{A_0}{A_t} \right]^q = \left[ \frac{X_{i,0}}{X_i} \right]^q \quad (5.65)$$

and applying the expression for  $X_i$  (eqn. 3.42):



$$F_{ca} = \frac{F_{ca,0}}{(1-sE)^{\frac{q}{v+1}}} \quad (5.66)$$

Equation 5.66 now becomes the boundary condition of the diffusion equation at  $\phi = 1$ .

Integration of the mass balance (eqn. 3.41) by using the expressions for  $X_i$  (eqn. 3.42) and  $F_{ca}$  (eqn. 5.66) gives:

$$\tau = \frac{(v+1) \left[ 1 - (1-sE)^{\frac{q+1}{v+1}} \right]}{(q+1) s F_{ca,0} X_{i,0}} \quad (5.67)$$

The relationship among  $F_{ca,0}$ ,  $E$  and  $E'_i$  has to be derived from the numerical solutions; however, this was not investigated.



## CHAPTER VI

### EVALUATION OF EXPERIMENTAL DRYING CURVES

#### 6.1 Introduction.

To investigate the practical aspects of power law diffusion, drying experiments were carried out using aqueous maltodextrin solutions. In a vacuum drying apparatus samples with a slab geometry were dried isothermally; the weight of the sample as function of time was registered automatically by means of a micro-computer.

A slab geometry has some very important advantages over other geometries, viz.:

- the data reduction of the experimental drying curves is very simple, because the relative simple correlations derived for non-shrinking systems, may be applied irrespective of the degree of shrinkage;
- the exchange surface area is independent of the shrinkage behaviour and thus remains constant during an experiment;
- samples can be prepared easily and reproducibly;
- isothermal drying conditions can be created relatively easily.

This chapter deals first with some theoretical aspects with respect to the data reduction of slab drying experiments, viz. the calculation of concentration dependent diffusion coefficients according to Schoeber's method [23] and the description and prediction of drying curves. Next

the drying apparatus and the experimental procedure are described. Finally the results of the drying experiments are evaluated. Some relevant properties of maltodextrin are summarized in Appendix F.

## 6.2 Theoretical Aspects.

### 6.2.1 Determination of Diffusion Coefficients.

Schoeber [23] describes a method to derive diffusion coefficients from experimental Regular Regime (with  $m_1=0$ ) drying curves. By analyzing several concentration dependences of the diffusion coefficient he correlated  $Sh_d$  with  $d \ln F / d \ln(1-E)$ . He observed that a power law dependence showed a good "average" behaviour: irrespective of the concentration dependence of the diffusion coefficient,  $Sh_d$ -values deviate less than 15% from the  $Sh_d$ -values belonging to the power law dependence. Schoeber's method and its practical application are described below.

In Chapter IV the following expression (eqns. 4.45 and 4.46) for the flux parameter F was introduced:

$$F = \frac{1}{2} Sh_d D_r (\bar{m} - m_1) \quad (6.1)$$

in which,

$$D_r (\bar{m} - m_1) = \int_{m_1}^{\bar{m}} D_r dm \quad (6.2)$$

From equations 6.1 and 6.2 with  $m_1=0$  follows:

$$\int_0^{\bar{m}} D_r dm = \frac{2F}{Sh_d} \quad (6.3)$$

Differentiation of this equation yields:

$$D_r = \frac{d(2F/Sh_d)}{d\bar{m}} \quad \text{at } m=\bar{m} \quad (6.4)$$

and the equation for the calculation of the diffusion coefficient follows by applying the definitions of  $D_r$ ,  $F$  and  $\bar{m}$  (see §3.5):

$$D = \frac{1}{(\bar{\rho}_s)^2} \frac{2}{Sh_d} \frac{d(j_{m1}^s d_s R_s)}{d\bar{u}} \quad \text{at } u=\bar{u} \quad (6.5)$$

The value of  $Sh_d$  is found from the Regular Regime correlation between  $Sh_d$  and  $d \ln F / d \ln(1-E)$ . Assuming a power law concentration dependence of the diffusion coefficient, it follows from equations 5.13-5.16 for a slab geometry:

$$Sh_d = 4.935 + 2.456 \frac{a}{a+2} \quad (6.6)$$

where  $a$  is found from:

$$\frac{d \ln F}{d \ln(1-E)} = a+1 \quad (6.7)$$

Equation 6.5 can also be expressed in terms of the efficiency  $E$  as follows below.

From the mass balance over the slab follows ( $d_s R_s = \rho_{s0} R_0$ ):

$$j_{m1}^s d_s R_s = d_s^2 R_s^2 (u_0 - u_x) \frac{dE}{dt} \quad (6.8)$$

In case of perfect shrinkage behaviour ( $\sigma=1$ ) the relation between  $\bar{\rho}_s$  and E is given by:

$$\frac{\bar{\rho}_s}{\rho_{s0}} = \frac{1+v_0}{1+v_0(1-E)} \quad (6.9)$$

and from equations 6.5, 6.8 and 6.9 follows:

$$D = \frac{2R_0^2}{Sh_d} \left[ \frac{1+v_0(1-E)}{1+v_0} \right]^2 \frac{d\left(\frac{dE}{dt}\right)}{d(1-E)} \quad (6.10)$$

The mass balance can also be written as:

$$F = \frac{R_0^2}{D_0} \frac{dE}{dt} \quad (6.11)$$

so equation 6.7 becomes:

$$\frac{d \ln\left(\frac{dE}{dt}\right)}{d \ln(1-E)} = a+1 \quad (6.12)$$

Slight modification of Schoeber's method

If power law diffusion is assumed, then:

$$F = \frac{1}{2} \frac{Sh_d}{a+1} (1-E)^{a+1} \quad (6.13)$$

From equations 6.11 and 6.13 follows:

$$\frac{dE}{dt} = \frac{1}{2} \frac{Sh_d}{a+1} \frac{D_0}{R_0^2} (1-E)^{a+1} \quad (6.14)$$

and from equations 6.10 and 6.14 can be derived, that the diffusion coefficient can also be calculated by using:

$$D = D_0 \left[ \frac{1+v_0(1-E)}{1+v_0} \right]^2 (1-E)^a \quad (6.15)$$

in which  $a$  and  $D_0$  are found by linear regression over small intervals of  $\ln(dE/dt)$  versus  $\ln(1-E)$ :

$$\ln\left(\frac{dE}{dt}\right) = \ln\left[\frac{1}{2} \frac{Sh_d}{a+1} \frac{D_0}{R_0^2}\right] + (a+1)\ln(1-E) \quad (6.16)$$

### 6.2.2 Description of Isothermal Drying Curves.

During drying of slabs with a high initial flux three drying stages can be distinguished:

- a relatively short period with a nearly constant surface moisture activity and thus a nearly constant drying flux;
- a Penetration Period with a nearly constant surface concentration ( $m_i \approx 0$ ), followed by
- a Regular Regime ( $m_i \approx 0$ )

In case of power law diffusion the isothermal drying of a material can be described fully by only two parameters, accounting for the concentration dependence of the diffusion coefficient, either  $a$  and  $b$  or  $a$  and  $D_0$ .

These parameters can be derived from the Regular Regime drying curve by means of linear regression of  $\ln(dE/dt)$  versus  $\ln(1-E)$  according to equation 6.16, however, now regression should be performed over all data points of the Regular Regime.

For high initial drying fluxes the Penetration Period should be

evaluated by using the expression (see Chapter V):

$$t = \frac{1}{2} \frac{R_0^2}{G_0 D_0} E^2 \quad (6.17)$$

in which,

$$G_0 = \frac{2}{\pi} \left[ \frac{1.42}{a+1.42} \right]^{1.98} \quad (6.18)$$

and from equation 5.1:

$$D_0 = \frac{b(u_0 - u_*)^a}{\rho_{s0}^2} \quad (6.19)$$

Data reduction by linear regression of  $t$  versus  $E^2$  gives a value for the slope  $R_0^2/(2G_0 D_0)$ . Finding  $a$  and  $b$  from the slope, in combination with equations 6.18 and 6.19, requires at least two experiments at different initial concentrations  $u_0$ .

### 6.3 Experimental.

#### 6.3.1 Description of Drying Apparatus.

As we have seen in §6.2 the data reduction of the experiments requires the first and second derivatives of the drying curves. Therefore the measurement of the weight should be as free of noise as possible and many data points of weight versus time should be acquired. For this reason the drying is performed under vacuum conditions (no noisy drag forces caused by a streaming gas) and the experimental set-up is fully automated by means of a micro-computer.

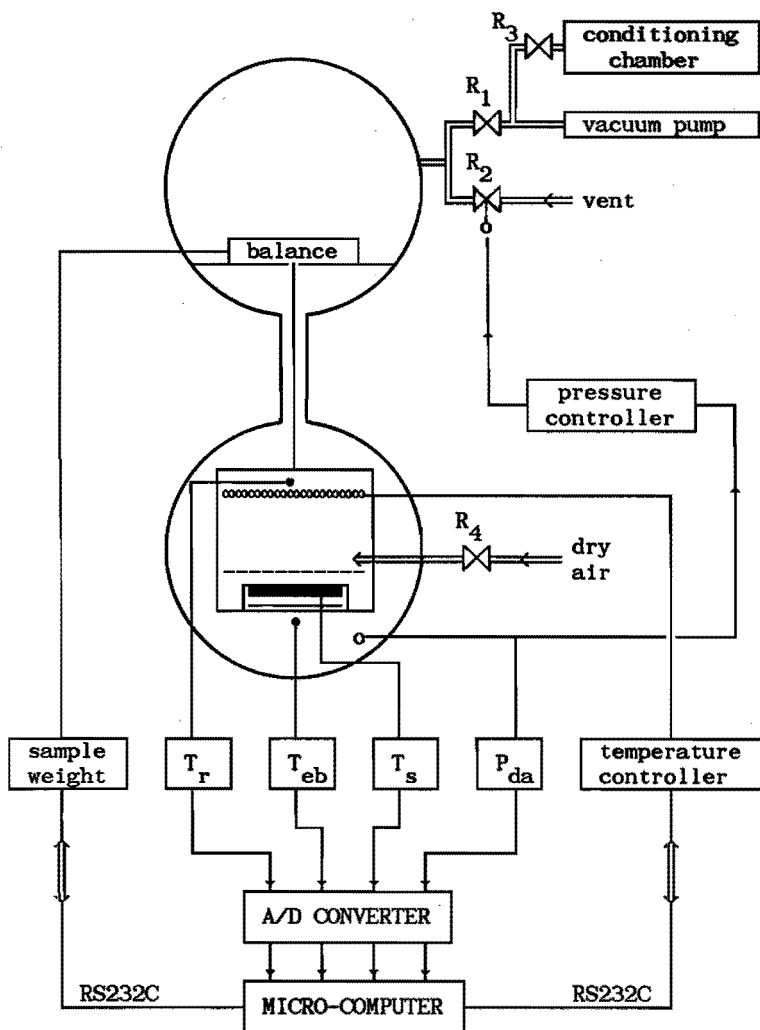


The drying apparatus (Figure 6.1) consists of two horizontal cylindrical chambers, one on top of the other. The chambers are connected with each other by means of a tube and are accessible at the front. The diameter and the depth of each chamber are 50 cm. The sample holder in the lower chamber is connected to an electronic precision balance (resolution 1 mg) in the upper chamber.

The temperature of the sample is kept constant by an electrically heated radiation wire, placed about 10 cm above the sample holder. The temperature of the sample layer is measured with a thermocouple (type T, Cu/CuNi). The power supplied to the radiation wire is regulated by a temperature controller.

To dilute the evaporated moisture ( $\rho'_{mo} \approx 0$ ) clean and dry air ( $R_4$ ) is blown as evenly as possible over the sample. In order to avoid uneven drying of the slab a sieve plate is placed between the sample and the air stream. The distance between sieve plate and sample is about 1 cm. This space may be considered as a diffusional resistance. The moisture diffusion coefficient in the gas phase is inversely proportional to the absolute pressure [1]. The initial rate of drying depends on the slab temperature, absolute pressure in the chambers and the distance between sample holder and sieve plate. The absolute pressure in chambers is kept constant by means of a pressure transmitter and a pressure controller, which activates the servo motor of a needle valve ( $R_2$ ). The sample holder is made of PTFE, which hardly takes up any moisture. Moreover, PTFE is a good heat insulator, which favours a uniform temperature distribution in the slab. The sample holder has a cylindrical shape with diameter 6 cm and depth 2.5 mm.

For data-acquisition the digital weight balance, the temperature controller, the pressure controller and several thermocouples are connected to a microcomputer. During a drying experiment the following



**Figure 6.1** Vacuum drying apparatus

■ = sample; --- = sieve plate;

○○○○ = radiation source;  $T_s$  = sample temperature;

$T_{eb}$  = temperature external bottom sample holder;

$T_r$  = temperature of radiation source;

$P_{da}$  = pressure in drying apparatus.

physical quantities are automatically recorded:

- the sample weight;
- the temperature of the slab;
- the temperature in the neighbourhood of the radiation wire; this temperature is a rough indication for the activity of the radiation source;
- the temperature of the external bottom of the sample holder (to ascertain a uniform temperature in the sample);
- the absolute pressure in the chambers (to ascertain constant external drying conditions).

### 6.3.2 Experimental Procedure.

#### Sample preparation.

To avoid internal circulations inside the slab, a gel of the aqueous maltodextrin solutions is prepared by adding a small amount of agar-agar (1 wt% on water basis). A sample layer is obtained by injecting the warm solution (50 °C) into the sample holder via a cover with two holes in it (Figure 6.2).

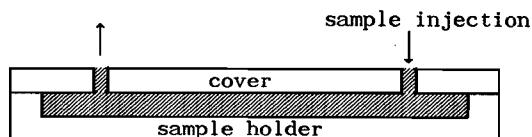
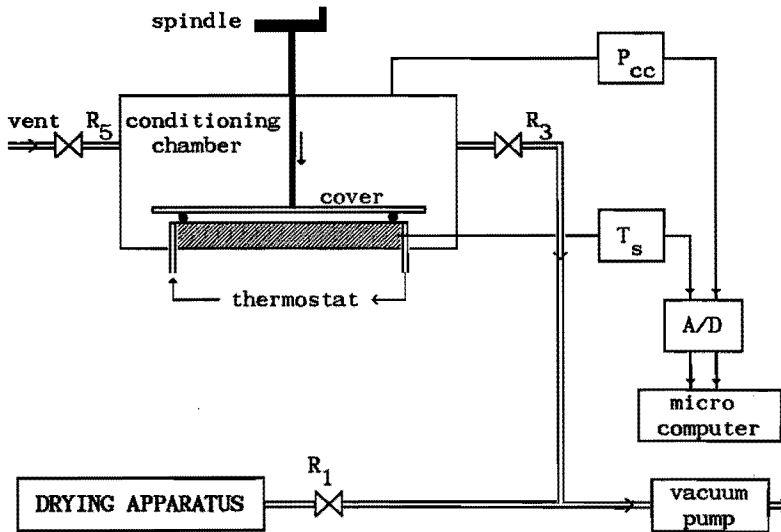


Figure 6.2 Preparation of a gelled layer.

After gelation at a lower temperature (e.g. 0°C), the cover is shifted away and a perfectly smooth slab is obtained. Immediately the sample holder is closed with a stainless steel cover plate provided with a "O" ring.

Sample conditioning.

It takes about three minutes to reach the desired pressure (about  $12000 \text{ N/m}^2$ ) in the drying chamber. To obtain a well-defined starting point it must be avoided, that during this period the sample starts drying. Therefore the sample, with a cover over it, is first placed in a separate conditioning chamber (Figure 6.3).



**Figure 6.3** Conditioning chamber

▨ = double-walled heat exchanger, in which sample holder is placed;

P<sub>cc</sub> = chamber pressure; T<sub>s</sub> = sample temperature.

At reaching the desired pressure (P<sub>cc</sub>) and temperature (T<sub>s</sub>) of the sample the cover plate with "O"-ring is pressed firmly to the sample holder by means of a spindle and then the chamber is brought to atmospheric pressure again.

Start of drying experiment.

Because of the underpressure in the small space between cover plate and sample surface, the sample holder will "stick" to the cover plate. In this condition the sample holder is placed in the drying chamber, whereby the cover plate is resting on two bars (Figure 6.4).

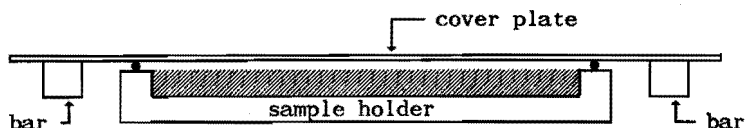


Figure 6.4 Position of sample holder just before the start of a drying experiment (▨ = sample layer)

The drying chamber is closed and the pressure is brought to the desired value. A short time before the end pressure is reached, the sample holder releases from the cover plate and falls on the digital electronic weight balance. The sudden change of the balance signal is registered by the micro-computer and interpreted as the start of the experiment; at the same time the balance is zeroed and a motor is activated to pull away the cover plate.

During drying experiment.

About 1000 times during an experiment the computer samples the following data: time, signal of the weight balance, signals of thermocouples (sample, external bottom of sample holder, radiation source) and signal of pressure transmitter.

End of drying experiment.

The experiments are aborted if the drying efficiency is at least 0.95. The duration of an experiment varies from several hours to one day.

### Representation of results.

From the momentary, initial and final weight of the sample the drying efficiency  $E$  can be calculated quite simply. For the evaluation of the drying experiment (§6.2) also  $dE/dt$  is required. To obtain sufficiently accurate values for  $dE/dt$  the increase of the efficiency during a time interval should be large enough. Therefore a selection of about 100 time intervals from all collected data has been made; during the selected time intervals the increase of the efficiency is about 0.007-0.01.

In Table 6.1 a summary of the conditions used in the various drying experiments is given. The pertinent experimental data (efficiency  $E$  versus time  $t$ ) can be found in Appendix G.

**Table 6.1** Drying experiments of gelled maltodextrin/water layers ( $R_0=2.50$  mm,  $d_s=1610$  kg/m<sup>3</sup>)

exp. number	%wt of solid	temp. (°C)	pressure (N/m <sup>2</sup> )	initial weight of layer (g)	$\rho_{s0}$ (kg/m <sup>3</sup> )	$u_0$ (kg/kg)
5	29.4	41.4	12500	7.66	331	2.40
6	29.4	32.8	12450	7.84	331	2.40
7	29.4	26.4	12400	7.92	331	2.40
8	17.1	35.8	12400	7.51	183	4.85
9	17.1	26.7	12200	7.65	183	4.85

### 6.4 Results and Discussion.

From calculations, based on temperatures of the sample layer and of the external bottom of the sample holder, it could be confirmed that sample temperatures were uniform within  $\pm 0.2$  °C. It appeared, that the temperature change of a sample during a whole experiment was of

the same magnitude. The pressure in the drying chamber was constant within  $\pm 100 \text{ N/m}^2$ .

#### 6.4.1 Merging of drying curves.

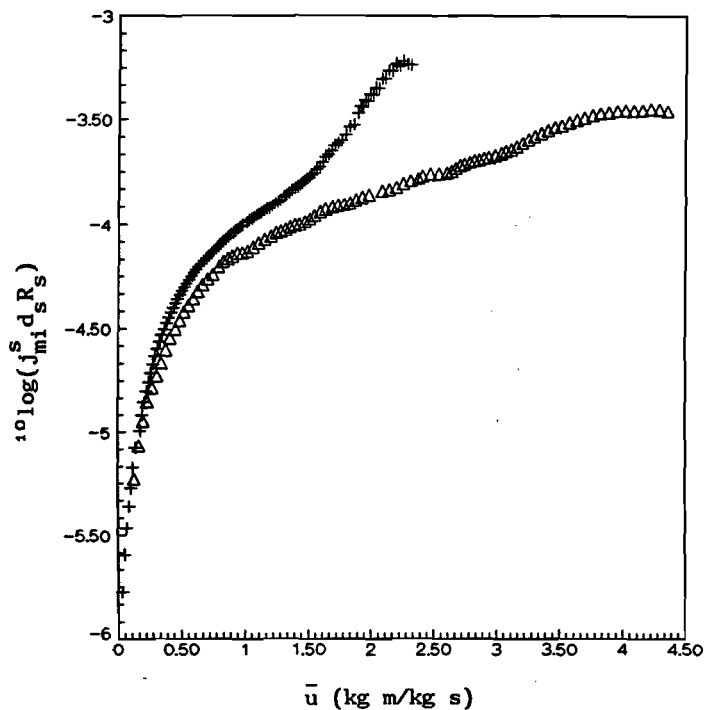
The relationship  $D$  versus  $u$  is a physical property of the material and so it has to be independent of the initial drying conditions.

Therefore it follows from equation 6.5 that curves  $j_{mi}^s d_s R_s$  versus  $\bar{u}$  for e.g. different initial moisture concentrations must merge in the Regular Regime.

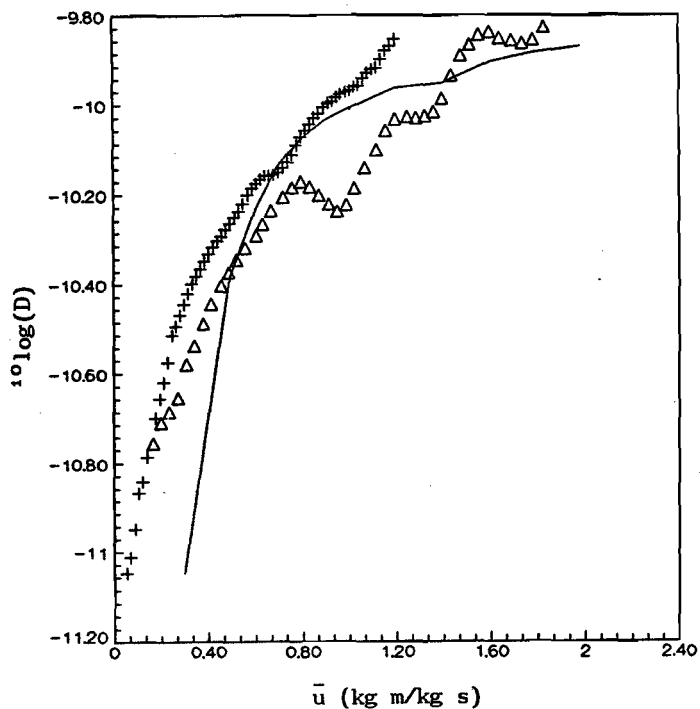
This conclusion has been verified experimentally by Schoeber [23] for aqueous glucose solutions and by Luyben et.al. [25-27] for a number of materials. From our investigation it appears, that also for aqueous maltodextrin solutions, at low concentrations the two drying curves coincide (Figure 6.5); from this merging part of the drying curves the initial conditions can not be reconstructed. Actually, Figure 6.5 gives the experimental proof for the occurrence of Regular Regimes.

#### 6.4.2 Diffusion coefficients.

Diffusion coefficients can be calculated from the experimental data in two ways: according to equation 6.10 and according to equation 6.15. Both methods have been used and they both require the relation of  $dE/dt$  versus  $(1-E)$ . To reduce noise and irregularities the first derivative  $dE/dt$  is smoothed by recalculating each data point as an average of its original value and four surrounding data points, two before and two after the original point; all points are weighted equally.



**Figure 6.5**  
 Merging of drying curves  
 with different initial  
 conditions.  
 + experiment 7  
 \Delta experiment 9



**Figure 6.6**  
 Diffusion coefficient of  
 water in aqueous malto-  
 dextrin solution at 27 °C  
 + experiment 7  
 \Delta experiment 9  
 — Furuta et.al.



At each data point the second derivative in equation 6.10 and the  $a$  and  $D_0$  values in equations 6.12 and 6.16 are determined via linear regression of 9 data points, 4 points before and 4 points after the central data point; all points are weighted equally.

It appears that diffusion coefficients calculated according to equation 6.10 do not deviate significantly from those calculated with equation 6.15. Diffusion coefficients, derived from experiments 7 and 9 with the modified Schoeber's method (eqn. 6.15) are represented in Figure 6.6. The oscillations, especially in experiment 9, due to experimental noise and irregularities, are not fully eliminated by smoothing and regression techniques. The random errors, caused by these oscillations, are about  $\pm 10\%$ .

Systematic deviations are found between diffusion coefficients, which have been derived from experiments with different initial moisture contents. For example, in a large concentration range diffusion coefficients from experiment 7 ( $u_0=2.40$  kg m/kg s) are about 25% higher than those from experiment 9 ( $u_0=4.85$  kg m/kg s). Because experiment 9 starts at a higher initial moisture concentration, higher efficiencies are obtained at the same actual moisture concentration than in experiment 7; from this it is concluded that experiment 9 is more "regular" and will give more reliable diffusion coefficients than experiment 7.

At lower concentrations the two drying curves coincide and nearly the same diffusion coefficients are found. Deriving diffusion coefficients from one drying curve, as proposed by Schoeber [23], appears to be doubtful. Deriving diffusion coefficients from the merging parts of several drying curves with different initial moisture concentrations, as done by Luyben et.al.[25], appears to be a better approach.

Diffusion coefficients at low concentrations will be more reliable, if

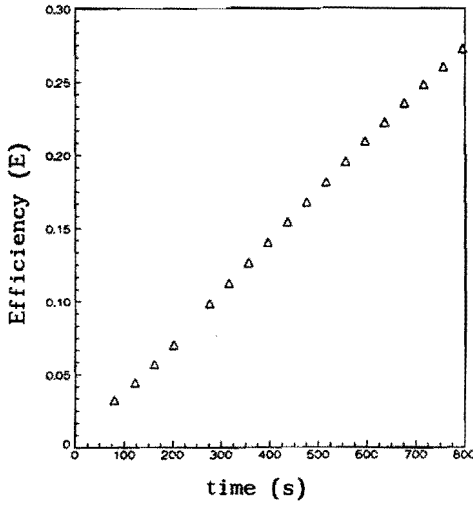
they are derived from experiments with low initial moisture concentrations. For example: at  $u=0.1$  (kg m/kg s) a drying efficiency of 0.975 is required if  $u_0=4$  and 0.9 is required if  $u_0=1$ ; in the first case relative errors in  $1-E$ , due to experimental inaccuracies, will be larger than in the second case.

Furuta et.al. [22] obtained diffusion coefficients of water in maltodextrin solutions from many desorption experiments. By applying low driving forces their concentration profiles were rather flat and the total change of the concentration during each single experiment was rather small. Therefore the diffusion coefficient could be assumed constant within those small concentration intervals. Their results (see Figure 6.6) are in fairly good agreement with ours, except for low concentrations ( $u < 0.3$  kg m/kg s), where large deviations exist.

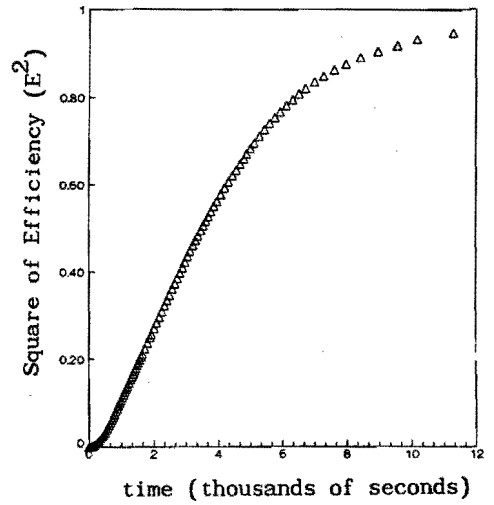
#### 6.4.3 Description of experimental drying curves.

A single drying experiment can be described quite well by means of the short-cut equations for power law diffusion (see Chapter V). This will be shown from experiment 9 as a typical example. In Figure 6.7 some characteristic relations, derived from this experiment, are represented.

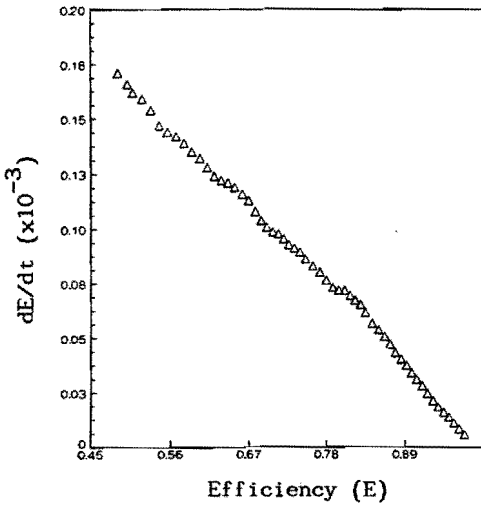
From a first examination of the Regular Regime drying curve it was estimated that  $a \approx 0$ . Therefore Regular Regime behaviour was assumed for  $E \geq 0.5$ . Linear regression of  $\ln(dE/dt)$  versus  $\ln(1-E)$ , including all data points in this range, gives a better value for  $a$  and thus a better estimate for the pertinent concentration range; after two or three iterations  $a = 0.075$  and  $D_0 = 1.00 \cdot 10^{-9} \text{ m}^2/\text{s}$  were found. Taking these values of  $a$  and  $D_0$ , the drying history can be reconstructed as follows.



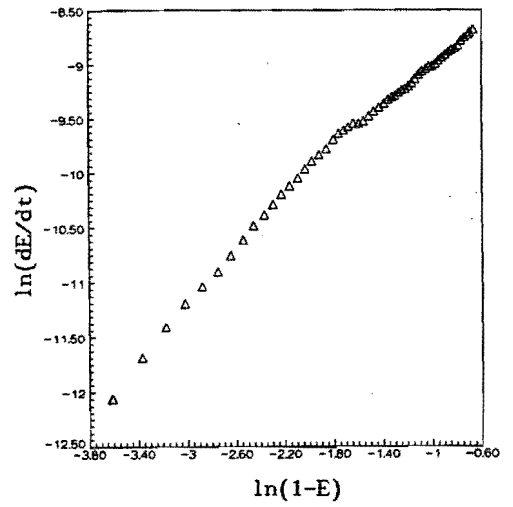
(a)



(b)



(c)



(d)

**Figure 6.7** Experiment 9 (see text)

(a)  $E$  versus  $t$

(b)  $E^2$  versus  $t$

(c)  $dE/dt$  versus  $E$

(d)  $\ln(dE/dt)$  versus  $\ln(1-E)$

Constant flux period

The imposed initial drying flux  $dE/dt = 3.50 \cdot 10^{-4} \text{ s}^{-1}$  is obtained by linear regression of  $E$  versus  $t$  in the arbitrarily chosen range  $0 \leq E \leq 0.2$ . Now the initial value of the flux parameter  $F_{ca} = 2.187$  follows from equation 6.11. The critical moisture concentration  $u_{cr} = 0.362 \text{ kg m/kg s}$  follows from the sorption-isotherm of maltodextrin/water (see Appendix F), assuming that the critical moisture activity  $a_{mi} = 0.9$ . From the definition of the efficiency follows for the critical surface efficiency  $E'_{i,cr} = 0.925$ . From equation 5.53 follows after iteration, that at the end of this period the average efficiency  $E_{ca} = 0.273$ .

Now the duration of the constant flux period can be calculated from the mass balance  $\tau_{ca} = E_{ca}/F_{ca} = 0.125$ . Applying the definition of  $\tau$  finally gives  $t_{ca} = 779$  seconds.

Penetration Period ( $m_i=0$ )

In case of the extreme situation that  $E'_{i,cr} = 1$ , it follows from equation 5.60 that  $E_{ca} = 0.325$  and  $t_{ca} = 928$  seconds. With respect to the total drying time the difference between 928 s and 779 s is small and therefore it is assumed now, that the transition period, in which the surface efficiency increases from the critical value ( $E'_i = E'_{i,cr}$ ) to the equilibrium value ( $E'_i = 1$  or  $m_i = 0$ ) is reached within a negligible time interval. This means that it is assumed, that the critical point curve ( $F_{ca}$  versus  $E_{ca}$  with  $m_{i,cr} = 0$ ) nearly coincides with the penetration curve ( $F$  versus  $E$  with  $m_i = 0$ ).

Because in this experiment the constant flux period contributes substantially to the total drying time, the Penetration Period should be calculated with:

$$t = t_{ca} + \frac{R_0^2}{2G_0D_0} (E^2 - E_{ca}^2) \quad (6.20)$$

in which,

$E_{ca} = 0.273$ ,  $t_{ca} = 779$  seconds and  $G_0 = 0.575$  (from eqn. 6.18).

### Transition

The transition from the Penetration Period to the Regular Regime occurs at  $E_T = 0.482$  (from eqn. 5.20).

### Regular Regime

For  $E > E_T$  the Regular Regime expression (from eqn. 5.18) reads:

$$t = t_T + \frac{2R_0^2}{Sh_d D_0} \frac{a+1}{a} \left[ \frac{1}{(1-E)^a} - \frac{1}{(1-E_T)^a} \right] \quad (6.21)$$

in which  $Sh_d = 5.024$  (from eqn. 6.6) and  $t_T = 1634$  s (from eqn. 6.20).

A comparison of measured and calculated drying times shows, that maximum deviations are about 10% (see Appendix G-9).

This result, which is typical for all drying experiments, proves that *the whole drying history can be reconstructed from experimental data of the Regular Regime.*

### Note

Though the initial value of the flux parameter is not very high in this case, it still ensures the occurrence of a Penetration Period with zero surface concentration ( $m_1 = 0$ ); higher initial values for  $F_{ca}$  can be achieved by higher temperatures, lower pressures in the drying chamber, thicker slabs and lower initial moisture contents (for obtaining lower values for  $D_0$ ); it should be noticed, however, that

thicker slabs and lower  $D_0$ -values will increase the duration of a drying experiment.

#### 6.4.4 Prediction of experimental drying curves.

In case of power law diffusion data reduction of all drying curves, irrespective of the initial concentration, should give one a-value and one b-value. However, it appears from our experiments, that power law diffusion does not apply strictly to solutions of maltodextrin/water (see also §6.4.6). Therefore a *semi-empirical approach* for the prediction of drying curves is proposed here. It appears that from drying curves at higher initial moisture contents the drying curves at lower initial moisture contents can be predicted. This will be illustrated by predicting drying experiment 7 ( $u_0 = 2.40 \text{ kg m/kg s}$ ) from drying experiment 9 ( $u_0 = 4.85 \text{ kg m/kg s}$ ).

The prediction method is based on the former observation, that the *Regular Regime drying curves contain all necessary information*. The values of a and  $D_0$  for the prediction of experiment 7 are derived from the Regular Regime of experiment 9. Assuming  $a = 0.075$  (so,  $E_T = 0.482$ ) the concentration range of the Regular Regime of experiment 7 is:  $0 \leq u \leq 1.16 \text{ kg m/kg s}$ . Linear regression of  $\ln(dE/dt)$  versus  $\ln(1-E)$  with all data points from experiment 9 in this range gives a better a-value and thus a better estimate of the relevant concentration range; linear regression is repeated, etc.. After only two iterations:  $a = 0.292$  and  $D_0 = 5.03 \cdot 10^{-10} \text{ m}^2/\text{s}$ . Similarly to the method, as described in §6.4.3, the drying times of experiment 7 are calculated. The maximum deviation between measured and calculated drying times appears to be 20% (see Appendix G-7).

The temperature dependence of drying curves, as stated by Schoeber [23,24], can be expressed with an Arrhenius-type relation:

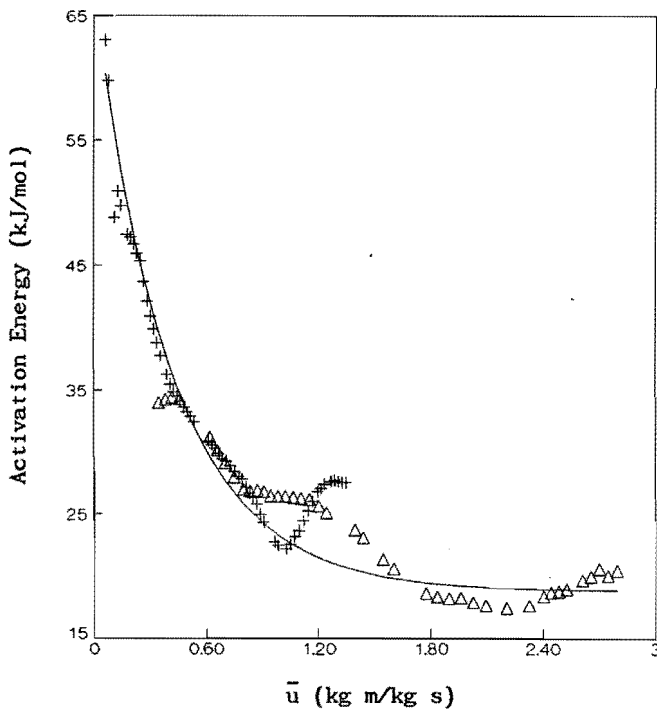
$$\left[\frac{dE}{dt}\right]_T = \left[\frac{dE}{dt}\right]_{T_1} \exp\left[-\frac{A_F}{R}\left(\frac{1}{T} - \frac{1}{T_1}\right)\right] \quad (6.22)$$

in which  $R = 8.314$  (J/mol  $^{\circ}\text{K}$ );

$T$  = absolute temperature ( $^{\circ}\text{K}$ );

$A_F$  = activation energy of the flux (Joule/mol).

The activation energy  $A_F$ , which depends on the moisture concentration, can be derived from at least two drying experiments with the same initial concentration, but with different temperatures.



**Figure 6.8**

Activation energy of  
drying flux.

+ from experiments 6 & 7

Δ from experiments 8 & 9

— equation 6.23

In Figure 6.8 the activation energies, derived from the Regular Regimes of two different sets of drying experiments (6+7 and 8+9) are represented. The concentration dependence of the activation energy  $A_F$  can be described very well by the following correlation, as proposed by Luyben [25]:

$$A_F = A \exp(-B u) + C \quad (6.23)$$

in which for maltodextrin/water the following values of the fitting parameters are found:  $A = 48$  kJ/mol;  $B = -2.40$  kg/kg,  $C = 18.8$  kJ/mol.

Isothermal drying curves at *different temperatures and different initial concentrations* can be predicted from experiment 9 as follows: first, the Regular Regime of experiment 9 is translated to the desired temperature by using equations 6.22 and 6.23.

second, the parameter  $a$  and  $D_0$  at this temperature are obtained by linear regression of  $\ln(dE/dt)$  versus  $\ln(1-E)$  in the concentration range of interest. Because  $a$  is not known beforehand, some iterations (mostly two) are required, to find the correct concentration range and the corresponding values of  $a$  and  $D_0$ .

third, the drying curve at the desired temperature level and desired initial concentration can be calculated similar to the method as described in §6.4.3.

The above procedure has been applied to predict the drying curves of experiments 5,6,(7) and 8 from the Regular Regime drying curve of experiment 9; the agreement of predicted and measured drying times is fairly good. Deviations between measured and calculated drying times vary from a few percent to 30% (see Appendix G).



#### 6.4.5 Prediction of Non-Isothermal Drying Curves.

At the desired initial moisture concentration, the Regular Regime curves are predicted at several temperature levels. Then by linear regression the best fitting  $a$ -value for all temperature levels is determined. Next the best fitting  $D_0$  value at each temperature level is determined. The temperature dependence of  $D_0$  is expressed with an Arrhenius type equation.

One single value of  $a$  is required to obtain temperature independent  $F$  versus  $E$  equations, so that the procedure analog to section 4.5 may be applied. The prediction of non-isothermal drying curves has not (yet) been verified experimentally in this study.

#### 6.4.6 Deviations from Power Law Diffusion.

The exponent  $a$  can also be derived from drying experiments at different initial concentrations via:

- correlating values of  $D_0$  with  $u_0$  according to equation 6.19;
- evaluation of values of  $G_0 D_0$ , found by linear regression of  $E^2$  versus  $t$  with data from the Penetration Period (see Figure 6.7), by means of equations 6.17-19.

In both cases negative  $a$ -values are obtained (Table 6.2), which strongly deviate from the foregoing observed values. This inconsistency of results means, that the power law relation:

$$D\rho_s^2 = b(u-u_*)^a \quad (6.24)$$

in fact does not apply to the whole concentration range of the system maltodextrin/water. At higher moisture concentrations the diffusion coefficient  $D$  is nearly independent of the concentration, whereas  $\rho_s^2$  will increase and thus  $D\rho_s^2$  will increase at a decreasing moisture concentration. At lower concentrations  $D$  will decrease more strongly

than the factor  $\rho_s^2$  increases and thus  $D\rho_s^2$  will decrease. Putting  $D\rho_s^2$  versus  $\rho_m/\rho_s$  will show a maximum, and such a dependence can not be expressed by a power law relation.

One may wonder now why the description and prediction of drying curves, based on the power law concept, gives such satisfying results. The answer to this question might be, that drying behaviour is fully controlled by the lower concentrations at the interface, whereas the concentration dependence of the diffusion coefficient at the higher concentrations in the drying material is of minor importance then. In the range of rate controlling concentrations apparently a power law dependence of the diffusion coefficient may be assumed. Because power law diffusion evidently does not apply to the whole concentration range it has to be concluded that the parameters  $a$  and  $D_0$  should be considered as fitting parameters, e.g.  $D_0$  is a fictive value of the diffusion coefficient at the initial concentration.

**Table 6.2**  $a$ -values calculated from drying experiments at different initial moisture concentrations.

exp	$u_0$ (kg/kg)	$G_0 D_0$ from PP ( $m^2/s$ )	$a$	$D_0$ from RR ( $m^2/s$ )	$a$
7	2.40	$2.37 \cdot 10^{-10}$	-0.61	$5.03 \cdot 10^{-10}$	-0.71
9	4.85	$5.04 \cdot 10^{-10}$		$1.00 \cdot 10^{-9}$	

#### 6.4.7 Conclusions.

Diffusion coefficients should be derived from the merging parts of several drying curves with different initial moisture concentrations.

The whole drying history of a sample can be reconstructed from experimental data of the Regular Regime only.

Drying curves at lower initial moisture concentrations can be predicted from the Regular Regime data of drying curves at higher initial moisture concentrations.

Power law diffusion does not apply strictly to maltodextrin/water solutions. Though drying curves can be described and predicted fairly well by means of the equations of the short-cut method, the model parameters  $a$  and  $D_0$  should be considered as fitting parameters with a limited physical meaning.



APPENDIX ATransformation of Diffusion Equation.

The diffusion equation for moisture transfer in bodies of standard geometries reads (§3.2):

Partial differential equation:

$$\frac{\partial \rho_m}{\partial t} = \frac{1}{r^v} \frac{\partial}{\partial r} (r^v n_m) \quad (\text{A.1})$$

Initial condition:

$$t = 0 \quad R_1 < r < R_2(0) \quad \rho_m = \rho_{m0} \quad (\text{A.2})$$

Boundary conditions:

$$t > 0 \quad r = R_1 \quad \frac{\partial(\rho_m/\rho_s)}{\partial r} = 0 \quad (\text{A.3})$$

$$r = R_2(t) \quad \rho_m = \rho_{mi}(t) \quad (\text{A.4})$$

or

$$-D\rho_s \frac{\partial(\rho_m/\rho_s)}{\partial r} = j_{mi}^s(t) \quad (\text{A.5})$$

By analogy with equation (A.1) the partial differential equation for mass transfer of the dissolved solids (s) is given by:

$$\frac{\partial \rho_s}{\partial t} = \frac{1}{r^v} \frac{\partial}{\partial r} (r^v n_s) \quad (\text{A.6})$$

Transforming the concentration.

For the transformation of the diffusion equation A.1, expressed in the volume based moisture concentration  $\rho_m$  ( $\text{kg/m}^3$ ), into a form with the concentration  $u$  ( $\text{kg m/kg s}$ ), the following two equations will be helpful:

$$\frac{\partial u}{\partial t} = \frac{\partial(\rho_m/\rho_s)}{\partial t} = \frac{1}{\rho_s} \frac{\partial \rho_m}{\partial t} - \frac{\rho_m}{\rho_s^2} \frac{\partial \rho_s}{\partial t} \quad (\text{A.7})$$

and from equations 2.13, 2.25 and  $u = \rho_m/\rho_s$ :

$$n_m = \rho_m v_s + j_m^s = u n_s - D \rho_s \frac{\partial u}{\partial r} \quad (\text{A.8})$$

Dividing equation A.1 by  $\rho_s$  and multiplying equation A.6 by  $\rho_m/\rho_s^2$  gives:

$$\frac{1}{\rho_s} \frac{\partial \rho_m}{\partial t} = - \frac{1}{\rho_s r^v} \frac{\partial}{\partial r} (r^v n_m) \quad (\text{A.9})$$

$$\frac{\rho_m}{\rho_s^2} \frac{\partial \rho_s}{\partial t} = - \frac{1}{\rho_s r^v} \frac{\rho_m}{\rho_s} \frac{\partial}{\partial r} (r^v n_s) \quad (\text{A.10})$$

Subtracting equations A.9 and A.10, making use of equation A.7 and eliminating  $n_m$  by means of equation A.8 results in:

$$\frac{\partial u}{\partial t} = \frac{1}{\rho_s r^v} \left[ \frac{\partial}{\partial r} (\rho_s r^v D \frac{\partial u}{\partial r}) - r^v n_s \frac{\partial u}{\partial r} \right] \quad (\text{A.11})$$

and with  $n_s = \rho_s v_s$  equation A.11 becomes:

$$\left( \frac{\partial u}{\partial t} \right)_r + v_s \left( \frac{\partial u}{\partial r} \right)_t = \frac{1}{\rho_s r^v} \frac{\partial}{\partial r} (\rho_s r^v D \frac{\partial u}{\partial r})_t \quad (\text{A.12})$$

### Transforming the space coordinate.

The solids based space coordinate  $z$  is defined as:

$$dz = \rho_s r^v dr \quad (\text{A.13})$$

or

$$z = \int_{R_1}^r \rho_s r^v dr \quad (\text{A.14})$$

and its maximum value:

$$z_{\max} = \int_{R_1}^{R_2} \rho_s r^v dr \quad (\text{A.15})$$

Transforming the right hand side of equation A.12 into a form, using the solids based space coordinate  $z$ , is quite simple, but for the left hand side this transformation is somewhat more demanding.

Consider  $u=u(t,z)$ , from which follows:

$$du = \left(\frac{\partial u}{\partial t}\right)_z dt + \left(\frac{\partial u}{\partial z}\right)_t dz \quad (\text{A.16})$$

and thus:

$$\left(\frac{\partial u}{\partial t}\right)_r = \left(\frac{\partial u}{\partial t}\right)_z + \left(\frac{\partial u}{\partial r} \frac{\partial r}{\partial z}\right)_t \left(\frac{\partial z}{\partial t}\right)_r \quad (\text{A.17})$$

From equation A.13:

$$\left(\frac{\partial r}{\partial z}\right)_t = \frac{1}{\rho_s r^v} \quad (\text{A.18})$$

From equation A.14:

$$\left(\frac{\partial z}{\partial t}\right)_r = \int_{R_1}^r \left(\frac{\partial \rho_s}{\partial t}\right)_r r^v dr \quad (\text{A.19})$$

Substitution of equation A.6 in A.19 and assuming a non moving internal boundary, it follows:

$$\left(\frac{\partial z}{\partial t}\right)_r = -r^v n_s = -r^v \rho_s v_s \quad (\text{A.20})$$

Equations A.17, A.18 and A.20 now give:

$$\left(\frac{\partial u}{\partial t}\right)_r = \left(\frac{\partial u}{\partial t}\right)_z - v_s \left(\frac{\partial u}{\partial r}\right)_t \quad (\text{A.21})$$

and equation A.12 written in the solids based space coordinate  $z$  finally reads:

$$\left(\frac{\partial u}{\partial t}\right)_z = \frac{\partial}{\partial z} (D \rho_s^2 r^{2v} \frac{\partial u}{\partial z})_t \quad (\text{A.22})$$

Making the partial differential equation dimensionless.

Introducing the dimensionless parameters  $m$ ,  $D_r$ ,  $\tau$  and  $\phi = z/z_{\max}$  (see Chapter III) into the partial differential equation A.22 gives:

$$\left(\frac{\partial m}{\partial \tau}\right)_\phi = \frac{\partial}{\partial \phi} (D_r X^2 \frac{\partial m}{\partial \phi})_\tau \quad (\text{A.23})$$

in which the dimensionless  $X$ -parameter is defined as:

$$X = \frac{r^v d_{s,ap} R_s}{z_{\max}} \quad (\text{A.24})$$

Working out the  $X$ -parameter.

Separation of variables in equation A.13 gives:

$$r^v dr = \frac{dz}{\rho_s} \quad (\text{A.25})$$

Integration between the limits  $r=R_1$  and  $r=r$  and substituting  $dz=z_{\max} d\phi$  gives the following expression:



$$r^v = \left[ R_1^{v+1} + (v+1)z_{\max} \int_0^{\phi} \frac{d\phi}{\rho_s} \right]^{\frac{v}{v+1}} \quad (\text{A.26})$$

For  $z_{\max}$  holds:

$$z_{\max} = \int_{R_1}^{R_2(t)} \rho_s r^v dr = \int_{R_1}^{R_1+R_s} d_{s,ap} r^v dr \quad (\text{A.27})$$

In fact equation A.27 is the mass balance of the solids; executing the integration of the most right member gives:

$$z_{\max} = \frac{d_{s,ap}}{v+1} \left[ (R_1+R_s)^{v+1} - R_1^{v+1} \right] \quad (\text{A.28})$$

From equations A.24, A.26, A.28 and the hollowness factor  $\lambda$  (eqn. 3.18) the following expression for the X-parameter can be derived:

$$X = (v+1) \frac{1-\lambda}{1-\lambda^{v+1}} \left[ \lambda^{v+1} + (1-\lambda^{v+1}) d_{s,ap} \int_0^{\phi} \frac{d\phi}{\rho_s} \right]^{\frac{v}{v+1}} \quad (\text{A.29})$$

From the shrinkage model follows (eqn. 3.26):

$$\frac{d_{s,ap}}{\rho_s} = 1 + \sigma v \quad (\text{A.30})$$

and the expression for the X-parameter now becomes:

$$X = (v+1) \frac{1-\lambda}{1-\lambda^{v+1}} \left[ \lambda^{v+1} + (1-\lambda^{v+1}) \int_0^{\phi} (1+\sigma v) d\phi \right]^{\frac{v}{v+1}} \quad (\text{A.31})$$

Transforming the initial and boundary conditions.

It will be clear that the initial condition reads:

$$\tau=0 \quad 0 \leq \phi \leq 1 \quad m = 1 \quad (\text{A.32})$$

For the external boundary condition the flux equation

$$j_{mi}^s = -D\rho_s \frac{\partial u}{\partial r} \quad (\text{A.33})$$

can rather easily be transformed into a dimensionless form via the intermediate expression:

$$j_{mi}^s = -D\rho_s^2 \frac{r^v}{z_{\max}} \frac{\partial u}{\partial \phi} \quad (\text{A.34})$$

and by next applying equation 3.14 for the flux-parameter and equation A.24 for the X-parameter, the dimensionless flux equation becomes:

$$F = -D_r X_i \frac{\partial m}{\partial \phi} \quad (\text{A.35})$$

where parameter  $X_i$  follows from equation A.31 by putting  $\phi=1$ .

After transformation the external boundary condition for a known surface concentration reads:

$$\tau > 0 \quad \phi = 1 \quad m = m_i \quad (\text{A.36})$$

or for a known surface flux:

$$\tau > 0 \quad \phi = 1 \quad -D_r X_i \frac{\partial m}{\partial \phi} = F \quad (\text{A.37})$$

In a similar way the internal boundary condition reads:

$$\tau > 0 \quad \phi = 0 \quad X \frac{\partial m}{\partial \phi} = 0 \quad (\text{A.38})$$

APPENDIX BAnalytical Solutions of Diffusion Equation with Constant Boundary Flux( $D_1=1, \sigma=0$ ).

The diffusion equation is given by (see §4.1):

Partial differential equation:

$$\frac{\partial m}{\partial \tau} = \frac{1}{[\lambda+(1-\lambda)y]^v} \frac{\partial}{\partial y} ([\lambda+(1-\lambda)y]^v \frac{\partial m}{\partial y}) \quad (\text{B.1})$$

Initial condition:

$$\tau = 0 \quad 0 \leq y \leq 1 \quad m = 1 \quad (\text{B.2})$$

Boundary conditions:

$$\tau > 0 \quad y = 0 \quad \frac{\partial m}{\partial y} = 0 \quad (\text{B.3})$$

$$y = 1 \quad - \frac{\partial m}{\partial y} = F_{ca} = \text{constant} \quad (\text{B.4})$$

For all geometries the solutions can be described by:

$$\frac{E'}{F_{ca}} = X_{i,\sigma=0} \left[ \tau + f + \sum_{k=1}^{\infty} C_k \frac{\exp(-\mu_k^2 \tau)}{\mu_k} \right] \quad (\text{B.5})$$

The functions  $f$  and  $C_k$  and the characteristic root equations are given in the following tables.

Table B.1 Massive systems			
geometry	f	$C_k$	$\mu_k$
slab	$\frac{1}{2} y^2 - \frac{1}{6}$	$\frac{2 \cos(\mu_k y)}{-\cos(\mu_k)}$	$\tan(\mu_k) = 0$ $\mu_k = k \cdot \pi$ $-\cos(\mu_k) = (-1)^{k+1}$
cylinder	$\frac{1}{4} y^2 - \frac{1}{8}$	$\frac{J_0(\mu_k y)}{-J_0(\mu_k)}$	$J_1(\mu_k) = 0$
sphere	$\frac{1}{6} y^2 - \frac{1}{10}$	$\frac{2}{3y} \frac{\sin(\mu_k y)}{-\sin(\mu_k)}$	$\tan(\mu_k) = \mu_k$

Table B.2 Hollow cylinder
$f = \frac{1}{8} \frac{(2x^2 - 3\lambda^2 - 1)(1 - \lambda^2) - 4\lambda^2 \{ (1 - \lambda^2) \ln(x) + \lambda^2 \ln(\lambda) \}}{(1 - \lambda)^3 (1 + \lambda)}$
$C_k = - (1 - \lambda^2) \frac{Z_0(\alpha_k \lambda, \alpha_k) \cdot Z_0(\alpha_k \lambda, \alpha_k x)}{Z_0^2(\alpha_k \lambda, \alpha_k) - Z_0^2(\alpha_k, \alpha_k)}$
<p>with :</p> $\alpha_k = \frac{\mu_k}{(1 - \lambda)}$ $x = \lambda + (1 - \lambda)y$ $Z_n(a, b) = Y_1(a)J_n(b) - J_1(a)Y_n(b)$ $Z_0(a, a) = \frac{-2}{\pi a}$
$\mu_k : Z_1\left(\frac{\lambda}{1 - \lambda} \mu_k, \frac{1}{1 - \lambda} \mu_k\right) = 0$

Table B.3 *Hollow sphere*

$$f = \frac{\lambda^3(-5y^3+15y^2+3y-3)+\lambda^2(15y^2+6y-9)+\lambda(15y^2-6y-3)+5y^3-3y}{30(1+\lambda+\lambda^2)(\lambda+(1-\lambda)y)}$$

$$C_k = \frac{2\mu_k^2}{3} \frac{g_k \cos(\mu_k)}{h_k \sin(\mu_k) \cos(\mu_k) + i_k} \frac{(1-\lambda) \sin(\mu_k y) + \mu_k \lambda \cos(\mu_k y)}{(\lambda + (1-\lambda)y)}$$

$$g_k = -(\lambda^2 + \lambda + 1) \cdot [(1-\lambda)^2 + \mu_k^2 \lambda^2]$$

$$h_k = \lambda^3 \mu_k^4 + \lambda(1-\lambda)^3 \mu_k^2 - (1-\lambda)^4$$

$$i_k = \mu_k \left[ \lambda^3 \mu_k^4 + \lambda(1-\lambda)^2 (1+\lambda) \mu_k^2 + (1-\lambda)^4 \right]$$

$$\mu_k : [(1-\lambda)^2 + \lambda \mu_k^2] \tan(\mu_k) - (1-\lambda)^2 \mu_k = 0$$

APPENDIX CAnalytical Solutions of Diffusion Equation with Constant Boundary Concentration ( $D_1=1, \sigma=0$ ).

The diffusion equation is given by (see §4.1):

Partial differential equation:

$$\frac{\partial m}{\partial \tau} = \frac{1}{[\lambda+(1-\lambda)y]^v} \frac{\partial}{\partial y} ([\lambda+(1-\lambda)y]^v \frac{\partial m}{\partial y}) \quad (\text{C.1})$$

Initial condition:

$$\tau = 0 \quad 0 \leq y \leq 1 \quad m = 1 \quad (\text{C.2})$$

Boundary conditions:

$$\tau > 0 \quad y = 0 \quad \frac{\partial m}{\partial y} = 0 \quad (\text{C.3})$$

$$y = 1 \quad m = m_i = 0 \quad (\text{C.4})$$

For all massive and hollow geometries the solutions can be described by the following relations:

$$m = \sum_{k=1}^{\infty} A_k \exp(-\mu_k^2 \tau) \quad (\text{C.5})$$

$$F = \sum_{k=1}^{\infty} B_k \exp(-\mu_k^2 \tau) \quad \text{with } B_k = - \left[ \frac{\partial A_k}{\partial y} \right]_{y=1} \quad (\text{C.6})$$

$$1-E = X_{i,\sigma=0} \sum_{k=1}^{\infty} B_k \frac{\exp(-\mu_k^2 \tau)}{\mu_k^2} \quad (\text{C.7})$$

The functions  $A_k$ ,  $B_k$  and the characteristic root equations are given in the tables below.

Table C.1 Massive geometries			
geometry	$A_k$	$B_k$	$\mu_k$
slab	$\frac{2}{\mu_k} \frac{\cos(\mu_k y)}{\sin(\mu_k)}$	2	$\cos(\mu_k) = 0$ $\mu_k = (2k-1) \frac{\pi}{2}$ $\sin(\mu_k) = (-1)^{k+1}$
cylinder	$\frac{2}{\mu_k} \frac{J_0(\mu_k y)}{J_1(\mu_k)}$	2	$J_0(\mu_k) = 0$
sphere	$\frac{2}{\mu_k y} \frac{\sin(\mu_k y)}{-\cos(\mu_k)}$	2	$\sin(\mu_k) = 0$ $\mu_k = k\pi$ $-\cos(\mu_k) = (-1)^{k+1}$

Table C.2 Hollow cylinder
$A_k = \pi \frac{J_1(\alpha_k \lambda) \cdot J_0(\alpha_k)}{J_1^2(\alpha_k \lambda) - J_0^2(\alpha_k)} \cdot Z_0(\alpha_k \lambda, \alpha_k x)$ <p>with <math>\alpha_k = \frac{\mu_k}{1-\lambda}</math>  <math>x = \lambda + (1-\lambda)y</math></p> $Z_n(a, b) = Y_1(a)J_n(b) - J_1(a)Y_n(b)$ $B_k = 2(1-\lambda) \frac{J_1^2(\alpha_k \lambda)}{J_1^2(\alpha_k \lambda) - J_0^2(\alpha_k)}$ <p><math>\mu_k : Z_0\left(\frac{\lambda}{1-\lambda} \mu_k, \frac{1}{1-\lambda} \mu_k\right) = 0</math></p>

Table C.3 *Hollow sphere*

$$A_k = \frac{2 \sin[\mu_k(1-y)]}{\mu_k - \sin(\mu_k) \cos(\mu_k) \lambda + (1-\lambda)y}$$

$$B_k = \frac{2\mu_k}{\mu_k - \sin(\mu_k) \cos(\mu_k)}$$

$$\mu_k : (1-\lambda) \tan(\mu_k) = -\lambda \mu_k$$



APPENDIX DSome Aspects of the Numerical Solution of the Diffusion Equation.

In general, the diffusion equation with  $D_r = m^a$  has to be solved by numerical methods. Only some important aspects of the numerical approach, which may cause serious problems, will be highlighted here. It appears that some of these problems can be dealt with by appropriate transformations of the diffusion equation; this will be illustrated with the generalized diffusion equation in  $\phi$ -coordinates.

Moving external boundary

The numerical calculation of partial differential equations requires a rectangular discretisation grid, in other words equidistant steps in time and space should be used. However, in discretizing the linear  $r$ -coordinate of a shrinking system the external boundary will move through the grid. This problem can be solved by transforming the partial differential equation (eqn. 4.1) with a normalized linear space coordinate:

$$y(t) = \frac{r-R_1}{R_{2,t}-R_1} \quad (D.1)$$

During every time step  $R_{2,t}$  has to be calculated; this requires the integration of the moisture flux with time or the integration of the moisture concentration with space.

Transformation of the diffusion equation in  $\phi$ -coordinates seems to be a good alternative, because now a fixed external boundary is obtained. However, the integration now emerges in the  $X$ -parameter and the internal boundary condition for massive cylinders and spheres becomes

undetermined ( $\phi=0$  and  $\lambda=0 \rightarrow X=0 \rightarrow \partial m/\partial \phi=??$ ). This internal boundary problem also exists for non-shrinking systems. For cylinders and spheres with an infinite small value of  $\lambda$  the slope  $\partial m/\partial \phi$  is clearly zero. Therefore, it may be expected that replacing the internal boundary condition of massive systems with  $\partial m/\partial \phi=0$  will cause minor errors. Indeed, by comparing the numerical solutions calculated in  $\phi$ -coordinates with solutions calculated in  $y$ -coordinates no detectable errors could be traced.

### Singularity of the boundary flux

If the boundary concentration  $m_1$  takes on a zero value, the diffusion coefficient will also be zero. Because of a finite boundary flux, the concentration gradient at the external boundary will become infinite. This singularity causes loss of convergence and must be avoided. Also for high values of the exponent  $a$  and low values of  $m_1$  steep concentration gradients at the external boundary will result. Even this non-singular situation may lead to a non-acceptable loss of convergence. This problem can be eliminated by the following simple transformation of the concentration:  $\tilde{m} = m^{a+1}$ . The generalized diffusion equation then becomes:

Generalized partial differential equation:

$$\frac{\partial \tilde{m}}{\partial \tau} = (\tilde{m})^{\frac{a}{a+1}} \frac{\partial}{\partial \phi} (X^2 \frac{\partial \tilde{m}}{\partial \phi}) \quad (D.2)$$

Initial condition:

$$\tau = 0 \quad 0 \leq \phi < 1 \quad \tilde{m} = 1 \quad (D.3)$$

Boundary conditions:

$$\tau > 0 \quad \phi = 0 \quad X \frac{\partial \tilde{m}}{\partial \phi} = 0 \quad (D.4)$$

$$\phi = 1 \quad \tilde{m} = \tilde{m}_1 \quad (D.5)$$

$$\text{or} \quad -X_i \frac{\partial \tilde{m}}{\partial \phi} = F(a+1) \quad (D.6)$$

From the boundary condition at  $\Phi=1$  it can easily be seen that for finite values of  $F(a+1)$  the gradient of the transformed concentration  $\partial w/\partial\Phi$  will remain finite for any value of the boundary concentration. (The same holds for  $\partial w/\partial y$  and  $\partial w/\partial r$ ).

#### Inconsistency of initial and boundary condition

In modelling a real drying process it may happen that the idealized boundary condition is inconsistent with the idealized initial condition; e.g. a uniform initial concentration implies a zero boundary flux at  $t=0$ , whereas at  $t \downarrow 0$  the boundary condition may impose a non-zero flux. In these cases a discontinuity exists, where the external boundary condition and the initial condition should link up. This singularity tends to cause serious oscillations in the solutions and has to be dealt with.

Initially there is a drying period during which the concentration profiles are penetrating into the centre of the body. For this penetration period ( $0 \leq \tau \leq \tau_{pp}$ ), where the drying body behaves like an infinite thick body, the so-called Boltzmann transformation is most suitable to avoid this kind of singularity problems:

$$z = \sqrt{\tau} \frac{1-\Phi}{\sqrt{\tau_{pp}}} \quad (D.7)$$

Now the boundary  $\Phi=1$  is transformed to the origin  $z=0$ , whereas the initial condition ( $\tau=0$ ) is transformed to  $z=\infty$ . Thus, inconsistency of conditions no longer exists. At  $\tau=\tau_{pp}$  a switch back from  $z$ -coordinates to  $\Phi$ -coordinates is necessary.

Discretisation

At increasing values of  $\tau$  the concentration profiles change more slowly. To speed up the numerical calculations a gradually increasing time step is required. However, to determine the convergence of the numerical solutions, equidistant steps in space and time are required. This problem can be solved by transforming the time variable according to:  $\tau' = \sqrt{\tau}$ ; equidistant steps in  $\tau'$  yield growing steps in  $\tau$ , because  $\Delta\tau = 2\tau' \Delta\tau'$ .

The transformed diffusion equation is discretized by means of the Crank-Nicolson scheme. Discretisation yields a set of equations which is solved by using the Thomas-algorithm. The order of the discretisation errors appears to be:

$$(\Delta z)^2 \text{ and } (\Delta\tau')^2 \text{ for } 0 \leq \tau \leq \tau_{pp} ; (\Delta\phi)^2 \text{ and } (\Delta\tau')^2 \text{ for } \tau \geq \tau_{pp}$$

For non-shrinking systems with constant diffusion coefficient the numerical calculations show excellent agreement (deviations less than 0.1%) with the analytical solutions (Chapter IV). For power law diffusion excellent agreement exists (deviations less than 0.5%) with the calculations of Schoeber [23] and Liou [28] and with the calculations of regular regimes based on geometrically similar concentration profiles [23,44].

APPENDIX EThe Initial Value  $\tilde{G}_0$  for Power Law Diffusion with Constant Boundary Flux.

In case of a power law dependence of the diffusion coefficient with concentration, the  $\tilde{G}$ -parameter (see Table 5.1) is defined as:

$$\tilde{G} = \frac{\frac{\tilde{F}}{\tilde{E}_i} \tilde{E}}{X_i} = \frac{F(a+1)}{X_i} \frac{1-(1-E)^{a+1}}{[1-(1-E_i)^{a+1}]^2} \quad (\text{E.1})$$

and the initial value:

$$\tilde{G}_0 = \lim_{\tau \rightarrow 0} \tilde{G} = \frac{F(a+1)}{X_i} \lim_{\tau \rightarrow 0} \left[ \frac{1-(1-E)^{a+1}}{[1-(1-E_i)^{a+1}]^2} \right] \quad (\text{E.2})$$

If  $\tau \rightarrow 0$  then also  $E \rightarrow 0$  and  $E_i' \rightarrow 0$  and thus:

$$\begin{aligned} \lim_{\tau \rightarrow 0} \left[ \frac{1-(1-E)^{a+1}}{[1-(1-E_i)^{a+1}]^2} \right] &= \lim_{\tau \rightarrow 0} \left[ \frac{(a+1)E + \text{higher } E\text{-terms}}{[(a+1)E_i' + \text{higher } E_i'\text{-terms}]^2} \right] \\ &= \lim_{\tau \rightarrow 0} \left[ \frac{(a+1)E \cdot [1 + \text{higher } E\text{-terms}]}{(a+1)^2 E_i'^2 \cdot [1 + \text{higher } E_i'\text{-terms}]} \right] \\ &= \lim_{\tau \rightarrow 0} \left[ \frac{E}{(a+1)E_i'^2} \right] \end{aligned} \quad (\text{E.3})$$

From equations E.1, E.3, and 4.35:

$$\tilde{G}_0 = \lim_{\tau \rightarrow 0} \left[ \frac{F \cdot E}{X_i E_i'^2} \right] = \lim_{\tau \rightarrow 0} G = G_0 \quad (\text{E.4})$$

It is assumed that the drying process with a constant boundary flux starts with a homogeneous concentration profile. At extremely small drying times ( $\tau \rightarrow 0$ ), the moisture concentration at any place in the material, including the interface, has hardly changed from its initial value. So, the concentration dependence of the diffusion coefficient does not come to expression yet. Also the influence of shrinking, if present, will be negligible at this very first drying stage. For these reasons it can be concluded that, *irrespective of the concentration dependence of the diffusion coefficient and the shrinkage behaviour, in all cases the  $G_0$ -parameter takes on the same value.* So, the value obtained for non-shrinking systems with constant diffusivity (eqn. 4.36) also holds in general:

$$\tilde{G}_0 = G_0 = \frac{\pi}{4} \quad (E.5)$$

APPENDIX FSome Properties of Maltodextrin.

The maltodextrin (trade name: Paselli MD20) used in this study is the same as used by Kerkhof [36] and Rulkens [35].

The composition as specified by the supplier (AvéBé, Veendam, the Netherlands), is:

- 1.5 wt% glucose
- 4.5 wt% maltose
- 9.3 wt% tri-saccharides
- 6.0 wt% tetra-saccharides
- 4.5 wt% penta-saccharides
- 74.2 wt% poly-saccharides

The partial density of maltodextrin in aqueous solutions may be considered to be constant [36] and is  $1610 \text{ kg/m}^3$ .

The sorption-isotherm in the range  $a_w \leq 0.9$  can be described very well with the G.A.B-equation [43]:

$$\frac{u}{u_1} = \frac{C \cdot k a_w}{(1 - k a_w) \cdot (1 - k a_w + C \cdot k a_w)} \quad (\text{F.1})$$

in which,

$$u_1 = 0.06343 - 8.586 \cdot 10^{-4} \cdot (\theta - 20) \quad (\text{kg m/kg s}) \quad (\text{F.2})$$

$$k = 0.9132 + 3.839 \cdot 10^{-3} \cdot (\theta - 20) \quad (\text{F.3})$$

$$C_g = 6.394 + 2.558 \cdot (\theta - 20) \quad (\text{F.4})$$

with  $15^\circ\text{C} \leq \theta \leq 50^\circ\text{C}$

The maltodextrin, used by Furuta et.al. [22], is of a different origin and has a different composition. In their paper they present the following correlation for the diffusion coefficient of water in maltodextrin solutions at 35°C:

$${}^{10}\log(D_w^{35}) = \sum_{i=0}^8 \frac{d_i}{(1+u)^i} \quad (\text{F.5})$$

$$\begin{aligned} \text{with } d_0 &= -9.62029, & d_1 &= 3.75424, & d_2 &= -86.5335, \\ d_3 &= 704.872, & d_4 &= -2853.10, & d_5 &= 6354.49, \\ d_6 &= -7952.04, & d_7 &= 5245.81, & d_8 &= -1424.05 \end{aligned}$$

For the activation energy of the diffusion coefficient is given:

$$E_D = \sum_{i=0}^5 \frac{e_i \cdot 10^4}{(1+u)^i} \quad (\text{F.6})$$

$$\begin{aligned} \text{with } e_0 &= 3.32582, & e_1 &= -15.8667, & e_2 &= 151.217, \\ e_3 &= -443.608, & e_4 &= 481.664, & e_5 &= -146.387 \end{aligned}$$



## APPENDIX G-5

## Experiment 5: measured and calculated drying curves of gelled layer of maltodextrin/water.

Conditions:  $u_0 = 2.40 \text{ kg m/kg s}$        $\theta = 41.4^\circ\text{C}$   
 $P = 12500 \text{ N/m}^2$        $R_0 = 2.50 \text{ mm}$   
 Fitting parameters:  $a = -0.092$        $D_0 = 3.88\text{E-}10 \text{ m}^2/\text{s}$

constant flux	penetration period	regular regime
$F_{ca} = 21.563$	$G_0 = 0.727$	$Sh_d = 4.816$
$G_0 = 0.889$	$E_T = 0.524$	
$u_{cr} = 0.398 \text{ kg/kg}$	$t_T = 3058 \text{ sec.}$	
$E'_{i,cr} = 0.834$		
$E_{ca} = 0.029$	<i>(about 70% of all data points, used in</i>	
$t_{ca} = 22 \text{ sec.}$	<i>the evaluation, are represented below)</i>	

E	time (s)		% rel. error	E	time (s)		% rel. error
	meas.	calc.			meas.	calc.	
0.020	20	15	-23.9	0.562	2888	3527	22.1
0.028	26	21	-18.6	0.570	2968	3630	22.3
0.045	38	35	-9.1	0.585	3128	3836	22.6
0.077	78	78	-0.6	0.594	3216	3952	22.9
0.098	118	120	1.4	0.605	3344	4109	22.9
0.132	198	205	3.5	0.623	3542	4365	23.2
0.146	238	247	3.9	0.633	3662	4518	23.4
0.171	318	335	5.5	0.653	3904	4821	23.5
0.182	358	381	6.4	0.662	4024	4973	23.6
0.193	398	427	7.2	0.671	4136	5114	23.6
0.215	478	523	9.5	0.689	4386	5422	23.6
0.225	518	572	10.4	0.697	4506	5560	23.4
0.243	598	669	11.9	0.713	4746	5853	23.3
0.252	638	716	12.2	0.720	4866	6002	23.3
0.261	678	765	12.8	0.728	4986	6150	23.3
0.277	758	863	13.9	0.742	5226	6443	23.3
0.285	798	912	14.3	0.752	5396	6649	23.2
0.300	878	1010	15.0	0.769	5716	7023	22.9
0.307	918	1059	15.4	0.777	5876	7210	22.7
0.314	958	1108	15.7	0.785	6036	7396	22.5
0.342	1118	1306	16.8	0.801	6396	7802	22.0
0.354	1198	1405	17.3	0.809	6596	8016	21.5
0.379	1358	1603	18.0	0.824	6996	8443	20.7
0.391	1438	1704	18.5	0.832	7236	8695	20.2
0.403	1520	1809	19.0	0.839	7476	8925	19.4
0.421	1652	1975	19.5	0.855	7998	9429	17.9
0.435	1758	2109	19.9	0.862	8318	9706	16.7
0.455	1918	2309	20.4	0.877	8996	10276	14.2
0.465	1996	2406	20.5	0.884	9394	10586	12.7
0.474	2078	2506	20.6	0.892	9846	10923	10.9
0.493	2236	2708	21.1	0.907	10924	11643	6.6
0.502	2316	2806	21.2	0.914	11604	12032	3.7
0.520	2476	3005	21.4	0.928	13476	12913	-4.2
0.529	2568	3120	21.5	0.935	14768	13402	-9.2
0.538	2648	3221	21.7	0.942	16436	13944	-15.2

## APPENDIX G-6

Experiment 6: measured and calculated drying curves of gelled layer of maltodextrin/water.

Conditions:  $u_0 = 2.40 \text{ kg m/kg s}$        $\theta = 32.8 \text{ }^\circ\text{C}$   
 $P = 12450 \text{ N/m}^2$        $R_0 = 2.50 \text{ mm}$   
 Fitting parameters:  $a = 0.076$        $D_0 = 3.99\text{E-}10 \text{ m}^2/\text{s}$

constant flux	penetration period	regular regime
$F_{ca} = 5.951$	$G_0 = 0.574$	$Sh_d = 5.025$
$G_0 = 0.713$	$E_T = 0.482$	
$u_{cr} = 0.376 \text{ kg/kg}$	$t_T = 3291 \text{ sec.}$	
$E'_{i,cr} = 0.8431$		
$E_{ca} = 0.0823$	(about 65% of all data points, used in	
$t_{ca} = 217 \text{ sec.}$	the evaluation, are represented below)	

E	time (s)		% rel. error	E	time (s)		% rel. error
	meas.	calc.			meas.	calc.	
0.055	160	145	-9.5	0.597	4032	5083	26.1
0.074	192	196	2.0	0.606	4144	5242	26.5
0.092	240	241	0.4	0.614	4272	5397	26.3
0.124	320	334	4.2	0.629	4496	5686	26.5
0.148	400	423	5.7	0.645	4736	5999	26.7
0.160	432	475	9.9	0.652	4848	6149	26.8
0.181	512	572	11.7	0.669	5136	6519	26.9
0.192	560	626	11.7	0.676	5248	6679	27.3
0.210	640	725	13.3	0.695	5568	7109	27.7
0.232	736	857	16.4	0.712	5888	7546	28.2
0.239	768	906	17.9	0.721	6048	7764	28.4
0.255	848	1011	19.2	0.737	6368	8198	28.7
0.269	928	1116	20.2	0.752	6688	8636	29.1
0.277	976	1171	20.0	0.759	6848	8855	29.3
0.304	1136	1383	21.7	0.776	7248	9405	29.8
0.316	1216	1486	22.2	0.784	7456	9677	29.8
0.339	1376	1694	23.1	0.799	7856	10224	30.1
0.361	1536	1902	23.8	0.814	8304	10828	30.4
0.371	1616	2008	24.3	0.822	8544	11154	30.5
0.391	1776	2214	24.7	0.837	9056	11842	30.8
0.410	1936	2418	24.9	0.851	9584	12543	30.9
0.419	2016	2523	25.1	0.859	9904	12966	30.9
0.437	2176	2727	25.3	0.873	10560	13802	30.7
0.445	2256	2830	25.5	0.881	10928	14271	30.6
0.462	2416	3036	25.6	0.896	11776	15319	30.1
0.478	2576	3240	25.8	0.910	12768	16523	29.4
0.485	2656	3343	25.8	0.917	13376	17212	28.7
0.500	2816	3549	26.0	0.932	14784	18774	27.0
0.522	3056	3859	26.3	0.946	16736	20766	24.1
0.532	3168	4013	26.7	0.953	18016	21946	21.8
0.552	3424	4323	26.2	0.975	23344	27305	17.0
0.561	3536	4473	26.5	0.982	26032	30337	16.5
0.571	3664	4635	26.5	0.989	30688	35028	14.1

## APPENDIX G-7

Experiment 7: measured and calculated drying curves of gelled layer of maltodextrin/water.

Conditions:  $u_0 = 2.40 \text{ kg m/kg s}$   $\theta = 26.4 \text{ }^\circ\text{C}$   
 $P = 12400 \text{ N/m}^2$   $R_0 = 2.50 \text{ mm}$   
 Fitting parameters:  $a = 0.292$   $D_0 = 5.03\text{E-}10 \text{ m}^2/\text{s}$

constant flux	penetration period	regular regime
$F_{ca} = 3.898$	$G_0 = 0.440$	$Sh_d = 5.248$
$G_0 = 0.555$	$E_T = 0.436$	
$u_{cr} = 0.362 \text{ kg/kg}$	$t_T = 2854 \text{ sec.}$	
$E'_{i,cr} = 0.849$		
$E_{ca} = 0.082$	(about 60% of all data points, used in	
$t_{ca} = 262 \text{ sec.}$	the evaluation, are represented below)	

E	time (s)		% rel. error	E	time (s)		% rel. error
	meas.	calc.			meas.	calc.	
0.021	80	66	-18.1	0.566	4496	4823	7.3
0.047	160	150	-6.4	0.581	4736	5098	7.6
0.060	192	192	-0.1	0.589	4864	5236	7.6
0.085	272	268	-1.3	0.605	5136	5562	8.3
0.109	352	336	-4.6	0.620	5392	5863	8.7
0.121	400	373	-6.8	0.628	5552	6053	9.0
0.141	480	448	-6.6	0.645	5872	6435	9.6
0.151	512	487	-4.8	0.653	6032	6627	9.9
0.169	592	570	-3.6	0.669	6352	7014	10.4
0.186	672	655	-2.6	0.684	6688	7418	10.9
0.194	720	697	-3.1	0.692	6848	7618	11.2
0.209	800	786	-1.8	0.709	7248	8119	12.0
0.224	880	874	-0.7	0.725	7648	8624	12.8
0.237	960	963	0.3	0.733	7840	8878	13.2
0.262	1120	1138	1.6	0.748	8240	9392	14.0
0.274	1184	1225	3.4	0.755	8448	9653	14.3
0.296	1360	1403	3.1	0.771	8960	10311	15.1
0.316	1520	1574	3.5	0.786	9440	10936	15.8
0.325	1584	1662	4.9	0.794	9712	11305	16.4
0.345	1760	1847	4.9	0.809	10272	12042	17.2
0.362	1920	2018	5.1	0.824	10880	12851	18.1
0.370	2000	2104	5.2	0.832	11232	13318	18.6
0.386	2160	2273	5.2	0.846	11952	14274	19.4
0.394	2240	2359	5.3	0.854	12352	14797	19.8
0.409	2400	2529	5.4	0.868	13216	15928	20.5
0.427	2608	2738	5.0	0.882	14240	17215	20.9
0.437	2720	2863	5.3	0.890	14832	17963	21.1
0.454	2928	3088	5.5	0.904	16192	19625	21.2
0.473	3168	3349	5.7	0.918	17888	21622	20.9
0.483	3296	3488	5.8	0.926	18928	22793	20.4
0.501	3536	3750	6.0	0.949	23616	27915	18.2
0.510	3664	3882	5.9	0.956	25712	30140	17.2
0.527	3904	4149	6.3	0.971	31728	36692	15.6
0.543	4144	4415	6.5	0.985	41616	49962	20.1
0.551	4256	4548	6.9	0.992	54224	65396	20.6

APPENDIX G-8

Experiment 8: measured and calculated drying curves of gelled layer of maltodextrin/water.

Conditions:  $u_0 = 4.85 \text{ kg. m/kg s}$        $\theta = 35.8 \text{ }^\circ\text{C}$   
 $P = 12400 \text{ N/m}^2$        $R_0 = 2.50 \text{ mm}$   
 Fitting parameters:  $a = -0.087$        $D_0 = 9.27\text{E-}10 \text{ m}^2/\text{s}$

constant flux	penetration period	regular regime
$F_{ca} = 4.254$	$G_0 = 0.721$	$Sh_d = 4.824$
$G_0 = 0.882$	$E_T = 0.523$	
$u_{cr} = 0.383 \text{ kg/kg}$	$t_T = 1411 \text{ sec.}$	
$E'_{i,cr} = 0.921$		
$E_{ca} = 0.175$	(about 80% of all data points, used in	
$t_{ca} = 277 \text{ sec.}$	the evaluation, are represented below)	

E	time (s)		% rel. error	E	time (s)		% rel. error
	meas.	calc.			meas.	calc.	
0.021	38	33	-14.5	0.620	1988	1951	-1.9
0.049	78	78	0.1	0.632	2068	2030	-1.8
0.077	118	122	3.1	0.644	2148	2108	-1.9
0.101	156	161	3.0	0.656	2228	2188	-1.8
0.156	246	248	0.7	0.679	2388	2349	-1.6
0.178	286	283	-1.0	0.691	2468	2431	-1.5
0.199	326	319	-2.0	0.702	2548	2514	-1.3
0.218	366	356	-2.7	0.711	2616	2584	-1.2
0.254	446	436	-2.2	0.732	2786	2762	-0.9
0.267	478	468	-2.0	0.742	2866	2848	-0.6
0.287	530	519	-2.0	0.751	2940	2926	-0.5
0.302	570	560	-1.7	0.761	3030	3022	-0.3
0.329	650	641	-1.4	0.779	3190	3196	0.2
0.342	690	682	-1.1	0.788	3270	3285	0.5
0.355	730	724	-0.8	0.796	3350	3374	0.7
0.367	770	764	-0.7	0.804	3430	3465	1.0
0.390	850	845	-0.6	0.819	3586	3637	1.4
0.401	890	885	-0.6	0.827	3678	3739	1.6
0.411	930	925	-0.6	0.834	3758	3831	1.9
0.421	970	964	-0.6	0.844	3878	3968	2.3
0.441	1050	1043	-0.7	0.863	4122	4247	3.0
0.450	1090	1082	-0.8	0.872	4242	4384	3.3
0.459	1130	1120	-0.9	0.880	4362	4523	3.7
0.468	1170	1159	-0.9	0.887	4482	4656	3.9
0.486	1250	1237	-1.1	0.904	4802	5008	4.3
0.494	1290	1276	-1.1	0.913	5002	5215	4.3
0.502	1330	1313	-1.3	0.921	5182	5392	4.1
0.510	1370	1351	-1.4	0.928	5380	5592	3.9
0.528	1460	1439	-1.4	0.943	5908	6066	2.7
0.536	1500	1478	-1.5	0.951	6268	6358	1.4
0.543	1540	1517	-1.5	0.958	6694	6662	-0.5
0.550	1580	1555	-1.6	0.965	7272	7019	-3.5
0.573	1708	1679	-1.7	0.979	9794	8000	-18.3
0.580	1748	1719	-1.7	0.986	13260	8751	-34.0

## APPENDIX G-9

Experiment 9: measured and calculated drying curves of gelled layer of maltodextrin/water.

Conditions:  $u_0 = 4.85 \text{ kg m/kg s}$   $\theta = 26.7 \text{ }^\circ\text{C}$   
 $P = 12200 \text{ N/m}^2$   $R_0 = 2.50 \text{ mm}$   
 Fitting parameters:  $a = 0.075$   $D_0 = 1.00\text{E-}09 \text{ m}^2/\text{s}$

constant flux	penetration period	regular regime
$F_{ca} = 2.187$	$G_0 = 0.575$	$Sh_d = 5.024$
$G_0 = 0.714$	$E_T = 0.482$	
$u_{cr} = 0.362 \text{ kg/kg}$	$t_T = 1634 \text{ sec.}$	
$E'_{i,cr} = 0.925$		
$E_{ca} = 0.273$	(about 70% of all data points, used in	
$t_{ca} = 779 \text{ sec.}$	the evaluation, are represented below)	

E	time (s)		% rel. error	E	time (s)		% rel. error
	meas.	calc.			meas.	calc.	
0.032	80	92	15.1	0.589	2486	2292	-7.8
0.044	122	126	3.7	0.601	2566	2373	-7.5
0.070	202	200	-0.7	0.621	2726	2522	-7.5
0.099	276	281	1.8	0.631	2806	2596	-7.5
0.126	356	360	1.2	0.649	2966	2746	-7.4
0.140	396	400	1.0	0.660	3046	2836	-6.9
0.154	436	440	0.9	0.669	3126	2912	-6.8
0.182	516	518	0.3	0.687	3292	3071	-6.7
0.195	556	557	0.2	0.695	3372	3146	-6.7
0.222	636	633	-0.5	0.711	3532	3306	-6.4
0.235	676	671	-0.8	0.718	3612	3384	-6.3
0.260	756	741	-2.0	0.734	3772	3547	-6.0
0.272	796	776	-2.5	0.742	3864	3638	-5.8
0.284	836	810	-3.1	0.750	3956	3733	-5.6
0.305	916	880	-3.9	0.770	4196	3985	-5.0
0.316	956	917	-4.1	0.779	4316	4103	-4.9
0.336	1036	987	-4.8	0.797	4556	4352	-4.5
0.345	1076	1021	-5.1	0.805	4676	4478	-4.2
0.363	1156	1091	-5.6	0.820	4876	4719	-3.2
0.371	1192	1120	-6.0	0.827	4996	4849	-2.9
0.381	1236	1162	-6.0	0.835	5116	4980	-2.7
0.400	1326	1241	-6.4	0.853	5434	5337	-1.8
0.408	1366	1276	-6.6	0.861	5594	5523	-1.3
0.423	1446	1345	-7.0	0.876	5914	5878	-0.6
0.432	1486	1385	-6.8	0.884	6114	6090	-0.4
0.446	1566	1454	-7.1	0.899	6514	6518	0.1
0.453	1606	1488	-7.3	0.906	6714	6747	0.5
0.460	1646	1524	-7.4	0.914	6994	7034	0.6
0.487	1806	1665	-7.8	0.929	7606	7644	0.5
0.500	1886	1737	-7.9	0.936	7966	7988	0.3
0.520	2006	1852	-7.7	0.951	8950	8880	-0.8
0.532	2086	1923	-7.8	0.958	9550	9415	-1.4
0.556	2246	2071	-7.8	0.973	11276	10919	-3.2
0.567	2326	2144	-7.8	0.980	13642	11999	-12.0



## LIST OF SYMBOLS

a	exponent in power relation	
a	thermodynamic activity	
a	heat diffusivity	$\text{m}^2/\text{s}$
a	component in binary mixture	
A	mass exchanging area	$\text{m}^2$
A	activation energy	$\text{J/mol } ^\circ\text{K}$
A	fitting parameter in eqn. 6.23	$\text{J/mol}$
b	factor in power relation	$\text{kg}^2/\text{m}^4\text{s}$
b	component in binary mixture	
B	fitting parameter in eqn. 6.23	$\text{kg s}/\text{kg m}$
Bi	Biot number	
c	constant value	
c	molar concentration	$\text{mol}/\text{m}^3$
$c_p$	specific heat at constant pressure	$\text{J}/\text{kg } ^\circ\text{C}$
C	fitting parameter in eqn. 6.23	$\text{J}/\text{mol}$
d	(partial) density	$\text{kg}/\text{m}^3$
D	diffusion coefficient	$\text{m}^2/\text{s}$
$D_r$	reduced diffusion coefficient	
E	efficiency	
$\tilde{E}$	(see Table 5.1)	
F	dimensionless flux-parameter	
$\tilde{F}$	(see Table 5.1)	
G	help function for penetration processes	
$\tilde{G}$	(see Table 5.1)	
H	shrinkage factor	
J	diffusive mass flux	$\text{kg}/\text{m}^2\text{s}$
J	Bessel functions	
k	mass transfer coefficient	$\text{m}/\text{s}$
$k''$	modified mass transfer coefficient	$\text{kg m}^2/\text{s}$
l	characteristic dimension	$\text{m}$
L	latent heat of evaporation	$\text{J}/\text{kg}$
m	exponent in Sherwood correlation (gas phase)	
m	dimensionless mass concentration	
m	moisture	
$\tilde{m}$	(see Table 5.1)	
n	mass flux in stationary coordinate system	$\text{kg}/\text{m}^2\text{s}$

n	exponent in Sherwood correlations (gasphase)	
N	molar flux	$\text{mol/m}^2\text{s}$
Nu	Nusselt number	
P	pressure	$\text{N/m}^2$
Pr	Prandtl number	
q	heat flux	$\text{J/m}^2\text{s}$
q	parameter to account for influence of dynamic radius on mass transfer coefficient $k'$	
r	space coordinate	m
R	radius of cylinder and sphere or thickness of one-sided drying slab	m
Re	Reynolds number	
s	solid(s)	
s	modified shrinkage coefficient	
Sc	Schmidt number	
Sh	Sherwood number	
t	time	s
T	absolute temperature	$^{\circ}\text{K}$
u	solids based mass concentration	$\text{kg m/kg s}$
v	ratio of volume fractions of moisture and solids	
v	velocity	$\text{m/s}$
V	volume	$\text{m}^3$
x	molar fraction on total basis	$\text{mol/mol}$
X	dimensionless parameter	
y	dimensionless linear space coordinate	
z	solids based space coordinate	$\text{kg s/m}^{2-\nu}$

#### GREEK SYMBOLS

$\alpha$	heat transfer coefficient	$\text{J/m}^2\text{s } ^{\circ}\text{C}$
$\alpha$	correlation parameter in eqns. 4.37 and 5.60	
$\tilde{\alpha}$	correlation parameter in eqn. 5.52	
$\beta$	correlation parameter in eqns. 4.65 and 5.7	
$\gamma$	slope of linearized sorption-isotherm	
$\tilde{\gamma}$	correlation parameter in eqn. 5.52	
$\delta$	thickness film layer in gas phase	m
$\epsilon$	porosity	
$\epsilon$	(see Table 5.1)	
$\phi$	generalized space coordinate	
$\lambda$	hollowness factor	
$\lambda$	heat conductivity	$\text{J/ms } ^{\circ}\text{C}$



$\mu_k$	roots of characteristic equations	
$\nu$	geometry parameter	
$\nu$	kinematic viscosity	$\text{m}^2/\text{s}$
$\omega$	mass fraction on total basis	$\text{kg}/\text{kg}$
$\theta$	temperature	$^{\circ}\text{C}$
$\rho$	mass concentration	$\text{kg}/\text{m}^3$
$\sigma$	shrinkage coefficient	
$\tau$	dimensionless time	

### SUBSCRIPTS

a	air
ap	apparent
ca	constant activity
cr	critical
d	dispersed phase
D	diffusion
e	value at zero critical surface concentration
eff	effective
f	film
F	flux
H	heat
i	interface
i	component in mixture
m	moisture
M	mass
p	pure component
pp	penetration period
Q	transition point (Penetration Period/Regular Regime)
r	reduced value
s	solid
sat	saturated
T	transition point (Penetration Period/Regular Regime)
v	vapour
0	value at $t=0$
1	first root
1	internal radius
2	external radius
*	equilibrium value
#	reference value
$\infty$	bulk of gas phase

### SUPERSCRIPTS

-	average value
$\square$	volume average value
o	molar average value
s	solid
'	gas phase
'	local value
~	(see Table 5.1)



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#### Stelling 1

Voor systemen, waarin de diffusiecoëfficiënt volgens een machtsrelatie afhangt van de concentratie en de grensvlakconcentratie *ongelijk nul* is, neemt het kengetal van Sherwood (betrokken op disperse fase) géén constante waarde aan gedurende het Regular Regime. Uit eigen onderzoek blijkt, dat het kengetal van Sherwood als functie van de droogtijd dan een minimum vertoont.

*Llou, J.K., Proefschrift LUW, 1982*

*Reniers, P., Afstudeerverslag TUE, 1985*

#### Stelling 2

De berekening van droogcurven met een empirische correlatie wordt door Petersen ten onrechte voorspelling van droogcurven genoemd.

*Petersen, J.N., Drying Technology, 4 (1986);319-330*

#### Stelling 3

Het verlies van vluchtige aromacomponenten tijdens drogen vindt plaats gedurende het droogstadium, waarin de grensvlakconcentratie van de aromacomponent de waarde nul nog niet bereikt heeft. Bij rekenmodellen voor het aromatransport is de veel gehanteerde randvoorwaarde, dat reeds vanaf het begin de grensvlakconcentratie nul zou zijn, daarom onjuist.

*Kerkhof, P.J.A.M., Proefschrift TUE, 1975*

#### Stelling 4

Uit het vergelijken van stripexperimenten (alleen aromatransport) en droogexperimenten (water- én aromatransport) blijkt, dat in rekenmodellen voor het aromatransport tijdens drogen de kruisdiffusie-term niet verwaarloosd mag worden.

*de Boer, M., Afstudeerverslag TUE, 1986*

*Smits, J.H.P.M., Afstudeerverslag TUE, 1985*

#### Stelling 5

De wijze waarop Gupta et. al. op basis van beschikbare correlaties voor de warmteoverdracht een nieuwe correlatie samenstellen, leidt niet tot een beter begrip van het warmteoverdrachtsproces.

*Gupta et.al., Chem.Eng.Sci., 29 (1974), 839-843*

#### Stelling 6

De door Furuta et.al. berekende diffusiecoëfficiënten voor water in maltodextrine oplossingen zijn niet in overeenstemming met de door hen gepubliceerde droogcurven.

*Furuta et.al., J.Food Eng., 3 (1984); 169-186*

#### Stelling 7

Door Van de Lijn wordt niet onderkend, dat een kleine meetfout in de sorptie-isotherm zeer kritiek is bij het voorspellen van een explosie van een drogende holle druppel in een sproeidroger.

*van de Lijn, J., Proefschrift LUW 1976*

#### Stelling 8

Ook als de algebraïsche oplossing bekend is, kan numerieke discretisatie van differentiaal vergelijkingen rekentechnisch grote voordelen bieden.

*Bosch, M.L., Prakticumverslag TUE, 1985*

#### Stelling 9

Het gebruik van een interne standaard bij de analyse van vluchtige sporenc componenten (in bijv. sinaasappelsap) m.b.v. een destillatie/extractie methode kan tot een sterke vergroting van fouten leiden.

*van Spreuwel, M.C.W., Afstudeerverslag TUE 1984*

#### Stelling 10

De bewering van McCathren, dat het polijsten van klarinet-rieten voorkomt dat tijdens het spelen de capillairen in het riet gevuld raken met speeksel, is onjuist. Evenmin is de bewering juist dat door polijsten de ongewenste veroudering van het riet door enzymatische afbraak van de vezels wordt voorkomen.

*McCathren, D.E., The Instrumentalist, October 1985, 56-63*

#### Stelling 11

Een gezonde muzikale ontwikkeling van amateur Harmonie- en Fanfare-orkesten wordt vaak gehinderd door het traditionele verwachtingspatroon dat de gemeenschap heeft van deze orkesten.