The role of theoretical and mathematical modeling in scale-up

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THE ROLE OF THEORETICAL AND MATHEMATICAL MODELLING IN SCALE-UP

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ABSTRACT

Drying processes are complex because of the non-linearity of physical phenomena and of the distributed parameter character on many levels of consideration. At different scales the relative importance of phenomena will differ. A discussion of the level structure is presented and the concept of "critical grain size" is introduced. As a general criterion for the importance of particle diffusion resistance a modified Biot-number is defined. Theoretical models are treated for diffusional mass transfer, both rigorous and short-cut models. The integration of these models with approximating relations or models for specific drying processes is demonstrated for spray drying of foods and fluidized bed drying of bioproducts. Also a theoretical treatment of several quality aspects is given.

INTRODUCTION

Scaling up of drying processes is for many driers and products still largely a matter of empiry, supported by some processing rules of thumb, and a very sound knowledge of drier construction. Difficulties are especially met in scaling up processes for performance products, such as foods and bioproducts. In a previous paper [1], we have devoted attention to the interface and knowledge transfer problems between food producers,

equipment manufacturers and scientists. As basic problem we have the difficulty of integration of the three areas of expertise : product knowledge, equipment building and process insight. Another problem is the difficulty or impossibility for many companies to have all expertise within their organisation. As a partial remedy we see the incorporation of theoretical models into simulation packages, to be used on PC's, *along with sufficient insight into the backgrounds of the process*. It is the intention of this paper to treat some basic aspects of drying problems, and show by examples in spray drying and fluid-bed drying what additional advantages can be obtained from theoretical and mathematical models.

BASIC CONSIDERATIONS OF DRYING PROCESSES

Complicating factors

From a viewpoint of quantitative understanding drying processes have the following special difficulties :

- the physical processes are highly non - linear
- there are quite complicated exchange and interaction processes
- the dominating phenomena depend on drying conditions, and may change within the course of drying.

The *non - linearity* is illustrated in Figs. 1 - 3. In Fig.1 the *saturated water vapour concentration* $\rho'_{w,sat}$ is given versus the temperature. As it is related to the water vapour pressure by :

$$\rho'_{w,sat} = \frac{p_w^s M_w}{R T} \quad (1-1)$$

it varies very strongly with temperature. The second *non - linearity* is the *water vapour sorption isotherm*, as illustrated in Fig. 2 for aqueous maltodextrin solutions. Since at the drying surface the actual water vapour concentration is given by :

$$\rho_w'^* = a_w \rho'_{w,sat} \quad (1-2)$$

the evaporation potential from the interface is strongly influenced by the water activity.

Fig.1. Saturated water vapour concentration as a function of temperature

Fig.2. Water vapour sorption isotherm of aqueous maltodextrin solutions

Fig.3. Molecular diffusion coefficient of water in skim - milk

which may be assumed to be in equilibrium with the moisture content at the product side of the interface.

A third non-linear factor is formed by the *transport properties inside the material*, as they depend on water concentration and temperature. An example is given in Fig. 3, in which the molecular diffusion coefficient of water in skim - milk is given vs. the water concentration for two temperatures. For other materials the governing transport phenomena may be more complicated, e.g. capillary flow and vapour diffusion. Shrinkage may lead to changes in surface area; this should be accounted for depending on the isotropy [2].

The *complexity of exchange processes* is illustrated in a simple example of an exchange scheme in Fig. 4 for convective driers. Elements of drying material may obtain heat directly from the world outside the drier by means of conduction or various forms of radiation, but will exchange mass and heat generally with their direct micro-environment, again by various transport phenomena. The micro - environment in turn will exchange mass and heat with a larger part of the drier through convection and turbulent mixing. The macro - environment exchanges heat and mass with the external world.

In a still largely simplified interaction scheme for the spray drying of liquid foods [1], as shown in Fig. 5, it is clear that next to exchange of heat and mass many more interactions are present. These may be of a micro-scale nature, e.g. drying rate and degradation reactions in a given droplet, but also of macro-scale nature, e.g. the outlet

Fig.4. Diagram of heat and mass transfer mechanisms in drying

temperature dependence on overall drying rate for a given chamber size and particle size distribution. In fact the latter leads to micro-macro scale interactions : the droplets see the local temperature and humidity, the air sees all droplets.

When looking at drying processes in this way one sees many variables; the large number of interrelations however reduces considerably the number of variables one can choose, *the number of degrees of freedom is very limited*. Putting in too many fixed desires for a design may even decrease the number of choosable variables below zero, meaning that the overspecified problem cannot be solved.

One of the basic roles of modelling is to include the significant interrelations and physical phenomena, and from this set make predictions for different circumstances.

Controlling regimes for the drying rate : the modified Biot - number

One of the key factors in drying processes is to describe the limiting or controlling mechanisms for the drying rate. We can do this in a quite general way for convection

Fig.5. Diagram of some influences in spray drying of foods

drying. Normally we start with a moisture content X_0 at which the water activity is still close to one. The rate of drying is then determined by the external heat and mass transfer coefficients and the local air temperature and humidity. The drying rate will start to fall once the water activity *at the interface* decreases considerably; a practical measure is a critical value of $a_w = 0.90$. Now we can locate our drying process between two extreme situations. In one situation, *slow drying*, the moisture inside has been able to diffuse fast enough, and so the water concentration inside the material is virtually flat. In that case the average moisture content X is equal to the surface moisture content, and the overall critical water content equals the limiting critical water content, following from the isotherm. In *fast drying, or sharp drying*, the diffusion of moisture inside cannot keep up with the external removal, and concentration profiles are built up inside the drying material. Consequently the average critical water content will then be higher than the limiting critical water content. We can expand this consideration to the falling rate period as well. When internal diffusion can keep up with external removal of water, the product water content remains homogeneously distributed, and the water activity at the surface decreases according to the sorption isotherm values corresponding to the average moisture content; the process is *externally controlled*. In sharp drying the surface water concentration drops off rapidly and it is the internal rate of diffusion which determines the drying rate : the process is now *internally controlled*.

As a yardstick for the controlling resistance we here propose to use a modified Biot - number, defined as :

$$Bi = \frac{k_{g,eff} L \rho_{wg}^s}{D_{eff} \rho_s X_{cr}^*} \quad (1-3)$$

Here $k_{g,eff}$ is the effective mass transfer coefficient [m/s], in which macroscopic effects have been included. For a fluid bed this means that the increase in humidity of the drying air upon passage has been taken into account. The dimension of the product particle is L [m], ρ_{wg}^s is the saturation vapour concentration [kg/m^3], D_{eff} is the effective diffusion coefficient [m^2/s], and ρ_s is the concentration of solids [kg/m^3]. A high value of Bi now indicates that in the falling rate period the main resistance lies inside the drying material, a low value that the drying rate will be externally controlled. This is illustrated in Fig. 6. We roughly place the boundary between the regions at $N_{Bi} = 1$. As an example we consider a droplet of 100 μm of a carbohydrate solution in a spray drier,

Fig.6. Location of dominating resistance as represented by the Biot number

with a wet bulb temperature of 45°C . Assuming $Sh_c = 2$, we get for the air side mass transfer coefficient $k_{g,\text{eff}} \approx 0.2 \text{ m/s}$. From Fig.1 follows that $\rho_{ws}^s = 0.07 \text{ kg/m}^3$, and from Fig. 3. a value of D_{eff} of about $1 \times 10^{-10} \text{ m}^2/\text{s}$. At 45 % solids we have roughly $\rho_s \approx 500 \text{ kg/m}^3$, and a typical value of X_{cr}^* is about 0.4 kg/kg ds . Thus follows for the Biot number $Bi = 70$. This means that diffusion limitation will be of great importance in this process. If we would have a granule with the same dimensions, which would be dried in a drum drier, the time-average mass transfer coefficient would be much smaller, say $k_{g,\text{eff}} = 1 \times 10^{-3} \text{ m/s}$, and the Bi-number would be equal to $Bi \approx 0.35$, indicating that the main resistance would lie in the air phase.

The scale of observation : the concept of "grain size"

There is one rule which is very important in scale-up in general : "you can never scale-up all phenomena and effects in a similar way". Therefore it is important to decide what are the dominating features. A second rule, especially useful for drying is : "The relative importance of influences may and probably will change with scale". The latter statement takes into account that in full-scale operation one wants to make efficient use of energy and of equipment volume and thus configuration, air/product ratios and flow patterns may differ considerably from pilot or laboratory situation. Thus drying rate, air

Fig.7. Different scales of observation for spray drying

temperature and humidity distribution, product particle size, flow and residence time (distribution) are in general scale-dependent; as a consequence also for many product quality aspects will hold the same.

Analogously to the considerations in mathematical physics we will have to look at drying processes on a number of levels of the scale of observation. In terms of modeling we have a multi-level, multi-distributed system. Our approach may have to vary between very "coarse-grained" (whole equipment) and extremely "fine-grained" (element within a particle), as illustrated in Fig. 7. The critical size of observation, or "critical grain size" is determined by the level of the dominating effects, either on drying rate or on product quality. If we have easy estimating methods available, the determination of this critical scale is simplified. In the previous section we estimated for a droplet of carbohydrate solution in a spray drier a value of the Biot number of typically 70. This means that in scaling up we will have to take diffusional limitations into account, and thus the critical grain size in this case is the droplet size. For systems in which Bi is very low, the critical grain size is that of a characteristic element of the drying air.

Of course the above is a bit simplified; one has to consider various aspects of the drying process, which may differ in the scale of importance. However the approach has the advantage that one is able to concentrate on sub-processes, without losing track of the

greater coherent process structure. In multi-disciplinary discussions about scale-up and product properties it may contribute enormously to the clarity if physical ideas are attached to the scale of consideration.

PRODUCT PROPERTIES

Generally speaking there is a host of product properties, of which a number are very product - specific, and others are somewhat more general. An attempt at a rough classification is the following :

piece properties : for discrete products like clay bricks, and gypsum boards very specific requirements are defined, like mechanical strength, absence of defects, and tolerances in dimensions.

intrinsic properties : these are properties in bulk products which are defined also for single particles or even small elements inside; examples are moisture content, aroma content, nutritional value, specific surface area for porous products.

extensive powder properties : some properties can only be related to many-particle systems, such as bulk density, flowability, wettability.

In general *piece properties* are very much dependent on the local drying history of elements of the dried units; unless we are drying at extremely low Biot numbers we will allways have differences between these histories in pilot or lab experiments and full-scale. Modelling including mechanical stresses and deformations may be required; the grain size is typically that of a small element within the drying material. Modelling is difficult and requires complex finite-element methods and the determination of constitutive properties. It may however lead to rules connecting local drying rates to occurrence of defects. Examples in this area are work on pasta by Andrieu et al. [3], and on clay by Ketelaars et al. in our laboratory [2]. The rules, combined with the increased insight, can be used in design of better lab experiments, and in analyzing local situations in full scale drying chambers.

Intrinsic properties are related to the average product properties by the following rules for averaging :

$$Y_{av} = \int_0^{\infty} \langle Y \rangle f(d_p) d(d_p) \quad (2-1)$$

in which $f(d_p)$ is the *particle size frequency distribution on dry mass base*. The quantity $\langle Y \rangle$ is the average of the property over the particle :

$$\langle Y \rangle = \frac{\int Y d(m_s)}{m_s} \quad (2-2)$$

in which Y is the local property defined on a dry mass base.

In lab or pilot experiments the local drying situation may differ from that in full scale, leading to different gradients inside a given particle with size d_p , leading to different $\langle Y \rangle$ for the same d_p . Moreover the particle size distribution will differ between scales, and thus also Y_{av} will be affected. Modelling may provide insight into the magnitude of these effects, and also provide directives towards measurement in various situations. A famous question in practice is why two identical driers in identical situations give different products; analysis of particle size distributions reveals in most cases that situations are not identical at all. Also this analysis may directly relate the effect of the different distributions to the considered properties.

Extensive powder properties are very important since they have a large impact on product handling and performance. Unfortunately they are the most difficult to describe quantitatively. From physical considerations however at least qualitative relations may be laid to surface area, particle size distribution, contact angle, surface composition. From modelling differences in situations may be understood, giving a better handle to improve properties. In our own laboratory we recently studied the spray - drying of high-fat food products and could explain properties like bulk density and free-fat content by approximating models.

OVERALL MASS AND ENTHALPY BALANCES

In any type of consideration balances must be made over the whole dryer. For the case of air drying without additional energy input we may write the general balances over the black box depicted in Fig. 8.

Fig.8. Black-box representation of drier

For solids we have :

$$P_{in} - P_{out} = \frac{d M_p}{d t} + V X_{s,fines} \quad (3-1)$$

in which P is the dry feed or product stream [kg solids/hr], M_p is the dry solids hold-up inside the drier [kg], V is the dry air stream [kg dry air/hr], and $X_{s,fines}$ is the fines load on dry basis in the outlet air.

For the moisture the balance reads :

$$P_{in} X_{in} - P_{out} X_{out} = \frac{d (M_p X_{av})}{d t} + V (H_{out} - H_{in}) + V X_{s,fines} X_{w,fines} \quad (3-2)$$

For the enthalpy the balance reads :

$$P_{in} j_{in} - P_{out} j_{out} = \frac{d (M_p j_{av})}{d t} + V (j_{out} - j_{in}) + V X_{s,fines} j_{fines} + \frac{d(J_{equip})}{d t} + \text{loss} \quad (3-3)$$

In this j is the enthalpy of the drying material, defined on dry solids base, and J_{equip} is the heat content of the equipment.

The enthalpy j can be expressed as :

$$j = c_{ps} + X_w c_{pw} \quad (3-4)$$

For the case of *continuous steady state drying, neglecting fines carry-over and heat losses*, we have $P_{\text{in}} = P_{\text{out}} = P$ and obtain :

$$P (X_{\text{in}} - X_{\text{out}}) = V (H_{\text{out}} - H_{\text{in}}) \quad (3-5)$$

$$P (j_{\text{in}} - j_{\text{out}}) = V (j_{\text{out}} - j_{\text{in}}) \quad (3-6)$$

For the case of a *fed - batch drier, again neglecting heat and fines losses*, we have :

$$\frac{d M_p}{d t} = P_{\text{in}} \quad (3-7)$$

$$\frac{d (M_p X_{\text{av}})}{d t} = P_{\text{in}} X_{\text{in}} - V (j_{\text{out}} - j_{\text{in}}) \quad (3-8)$$

$$\frac{d (M_p j_{\text{av}})}{d t} = P_{\text{in}} j_{\text{in}} - V (j_{\text{out}} - j_{\text{in}}) - \frac{d(J_{\text{equip}})}{d t} \quad (3-9)$$

The last term should in general be considered, especially in control models. However for many approximate studies we may neglect it. For a batch drier the above is easily simplified by putting $P_{\text{in}} = 0$, and M_p constant.

DISTRIBUTED MODELS FOR MASS TRANSFER AND DEGRADATION

The diffusion equation for water

It has been well established that in drying of many materials during a large part of the process the drying rate is determined by diffusional resistance inside the drying material. The transport mechanism may be molecular diffusion in an amorphous material,

such as in spray drying coffee extract, or a more complex simultaneous transport in small pores in a solid matrix as in the drying of structured materials such as clay. However also in the latter case one may with success also lump various phenomena into a model with an effective diffusion coefficient. In general the diffusion equation for such a system reads :

$$\frac{\partial \rho_w}{\partial t} = - \nabla \cdot (n_w) \quad (4-1)$$

with :

$$n_w = - D_w \nabla \rho_w + v^{\square} \rho_w \quad (4-2)$$

in which ρ_w is the water concentration [kg/m^3], n_w is the water flux with respect to a coordinate fixed to the drying specimen [$\text{kg/m}^2\text{s}$], D_w is the (effective) mutual diffusion coefficient of water and solids [m^2/s] and $v^{\square}$ is the volume-averaged velocity with respect to the chosen coordinate [m/s].

Often one can simplify the transport model to a simple geometry : an infinitely extended slab, drying from one side, an infinite cylinder or a (hollow) sphere. If then the coordinate is fixed with respect to the non-drying side of the slab or the centre of the cylinder or sphere respectively we obtain :

$$\frac{\partial \rho_w}{\partial t} = \frac{1}{r^v} \frac{\partial}{\partial r} \left[r^v \left(D_w \frac{\partial \rho_w}{\partial r} - v^{\square} \rho_w \right) \right] \quad (4-3)$$

in which v is a geometry factor : $v = 0$ for the slab, $v = 1$ for the cylinder, $v = 2$ for the sphere. If the system does not expand or shrink the volume-averaged velocity is equal to 0; this is also the case for systems with a shrinking boundary but without internal volume production or contraction (a slab in which the total shrinkage exactly balances the volume of water removed, or an isotropically drying totally filled sphere). In the case of an inflating droplet the velocity can be expressed in terms of the growth of the internal gas bubble :

$$v^{\square} r^2 = \frac{dR_g^3}{dt} \quad (4-4)$$

in which R_g is the radius of the gas bubble.

In order to complete the set we need the initial and boundary conditions. In general we have for the initial condition :

$$t = 0 \quad R_{in,0} \leq r \leq R_{out,0} \quad \rho_w = \rho_{w,0} \quad (4-5)$$

in which $R_{in,0}$ is the coordinate of the inner surface (which of course equals 0 for slabs, massive cylinders and solid spheres). In case $R_{in} = 0$ we have in general the no - flux condition as one of the boundary conditions :

$$t > 0 \quad r = 0 \quad \frac{\partial \rho_w}{\partial r} = 0 \quad (4-6a)$$

and for an inflating bubble approximately :

$$t > 0 \quad r = 0 \quad n_{w,b} \approx 0 \quad (4-6b)$$

in which $n_{w,b}$ is the water flux with respect to the bubble-liquid interface.

At the external interface we have the equality between the internal and the external water flux through the phase boundary :

$$j_{w,i} = -D_w \nabla \rho_w + (v^{\square} - v^i) \rho_w = k_{eff} (\rho'_{w,i} - \rho'_{w,b}) \quad (4-7)$$

in which k_{eff} is the effective mass transfer coefficient in the gas phase [m/s], and $\rho'_{w,i}$ and $\rho'_{w,b}$ are the water concentrations in the drying air at the interface and in the bulk respectively. The velocity of the interface is indicated by v^i , while $j_{w,i}$ stands for the flux of water with respect to the moving interface [kg/m²s].

At the interface we assume equilibrium between the two phases :

$$\rho'_{w,i} = A_{w,i} \rho'_{w,sat} = A_{w,i} \frac{P_{w,s} M_w}{R T} \quad (4-8)$$

in which $P_{w,s}$ is the saturation water vapour pressure [Pa], $\rho'_{w,sat}$ the corresponding water vapour concentration [kg/m³], M_w the molecular mass of water [kg/kmole], R the gas constant [J/kmole K], T the particle absolute temperature[K].

The water activity $A_{w,i}$ depends on the interfacial water concentration in the drying material and the temperature as described by the water vapour sorption isotherm :

Fig.9. Water concentration profiles inside a droplet of skim milk during spray drying

$$A_{w,i} = A_w (\rho_{w,i}, T) \quad (4-9)$$

In many practical situations the *limitation for heat transfer* lies in the drying air, leading to a virtually uniform temperature profile inside the drying material. We may then write an overall heat balance :

$$\alpha_{\text{eff}} (T_b - T) S = \frac{d}{dt} \left[m_s j + m_g i \right] + j_{w,i} i_v S \quad (4-10)$$

in which α_{eff} is the effective heat transfer coefficient [$\text{W/m}^2\text{K}$], T_b is the bulk temperature of the air [K], S is the surface area [m^2], m_s is the solids mass, and m_g is the included dry gas mass, and j and i stand for the enthalpies of the drying material *per kg of dry solids*, and i_v stands for the enthalpy of water vapour [J/kg].

The solution of the above equations has been subject of considerable work in the seventies [4-9]. In order to cope with the problems of moving boundaries coordinate transformations to solids-based coordinates were derived, leading to non-shrinking coordinates. Due to the concentration-dependence of the diffusion coefficient standard

numerical methods failed, and special numerical methods had to be developed. Systems studied in this way were the drying of slabs of maltodextrin solutions [4,5], of glucose solutions [6], of droplets of aqueous maltose solutions [7,8], and the drying of skim milk [9]. In Fig. 9 an example is given of the water concentration profile inside a solid droplet during spray drying.

Loss of volatile aroma components

Also this aspect has been subject of considerable study, both experimentally and theoretically. A recent overview is given by Coumans et al. [10]. The basic considerations for modelling are the simplification to a ternary system, consisting of water, dissolved solids and a trace aroma component. The aroma component is thought to be very volatile and not thermally degradable within the region of interest. Analogously to the water balance the diffusion equation for such an aroma component reads :

$$\frac{\partial \rho_a}{\partial t} = - \nabla \cdot (n_a) \quad (4-11)$$

In the flux equation for aroma however now both the effect of a gradient in aroma concentration and that in the water or solids concentration should be taken into account :

$$n_a = - D_{aa} \nabla \rho_a - D_{aw} \nabla \rho_w + v \rho_a \quad (4-12)$$

Both the straight aroma diffusion coefficient D_{aa} and the cross-diffusion coefficient D_{aw} are strongly dependent on the water concentration. At low water concentrations the diffusion coefficients become much lower than that of water; in a drying process thereby effectively a semi-permeable dry skin is formed through which aroma transport is virtually zero.

At the interface with the drying air because of the high volatility can be written :

$$t > 0 \quad r = R \quad \rho_a = 0 \quad (4-13)$$

The solution of the aroma diffusion equation can only be done simultaneously with that for the water diffusion. In a number of papers a detailed account is given of the results [6,8]. Systems considered until now have been layers and solid spheres. What was stated

for the solution of the water diffusion equation also holds for that of aroma : special numerical techniques had to be designed, and experimental data are very scarce.

Thermal degradation of heat-labile compounds

In a number of studies the thermal degradation of heat-labile components was investigated. For a large class of problems the degradation may be treated as first order reaction :

$$\frac{d \rho_i}{dt} = - k_r \rho_i \quad (4-14)$$

in which the degradation rate constant depends on the water concentration and temperature, for a given composition of the solids. For a number of heat-labile enzymes was found that the rate constant decreased sharply with decreasing water concentration and increased with temperature. This has been included in spray drying models [8,9]. A typical example could look like Fig. 10, as is used in modelling studies in courses by us. Also in a study of short-cut methods based on the power-law diffusion, Liou found short-cut methods to determine water concentration profiles inside the drying material, and integrated enzyme inactivation in his calculations. In the basic modelling it is assumed that the component, say enzyme, is immobile with respect to the dissolved solids phase. The diffusion equation is solved, and simultaneously the enzyme profile within the particle is calculated. This profile is then integrated over the particle to obtain the average rest activity.

In recent studies it has been shown that generally for such components as enzymes the above kinetics seem to hold [19,20]. However in the drying of micro-organisms such as yeasts or lactic acid bacteria a much larger degradation occurs than would be expected on the grounds of the separate inactivation kinetics integrated with the drying curve [12].

Regular Regimes and short-cut calculation methods

In his excellent thesis Schoeber developed a framework for short-cut calculation methods for desorption processes with concentration-dependent diffusion coefficients [13].

Fig.10. Typical dependence of degradation rate constant on water concentration and temperature

This was based on the Regular Regime observation : For a given set of boundary conditions the drying rate will after some time only depend on the actual moisture content, and the influence of the initial condition decreases to virtually zero. To describe a drying process he identified three stages : the *Constant Activity Period*, the *Penetration Period*, and the *Regular Regime*, and methods to calculate transition points between these regions. Furthermore he derived relationships between the concentration dependence of the diffusion coefficient and the Regular Regime part of the drying curve, thereby opening the way of determination of the diffusion coefficient from relatively simple drying experiments. The advantage of his methods was the generality of the treatment, which held for all kinds of concentration dependencies.

In further studies Liou and Coumans developed methods of also calculating the concentration profiles inside the material by short-cut methods, for the special case of a concentration dependence given by the power-law relation of the type $D = D_0 X_w^a$. Also they provided several simplified calculation methods for various geometries [11,12].

Fig.11. Drying curves for a layer of skim milk at 30 degs.C, at different initial water concentrations and fluxes, illustrating various drying regimes

In various treatments different definitions have been used of intermediate parameters, in the form of dimensionless groups. Here we have stripped the treatment to "bare-bone" variables; a certain loss of generality is thereby compensated by a more compact discussion.

Drying curves and Regular Regimes

As has been presented by Schoeber and others [13,14,15], a very convenient way of presenting a drying curve is that by plotting the flux parameter F vs. the average moisture content X on solids base.

The flux parameter is defined here as :

$$F = j_w R_s$$

(5-1)

in which j_w is the water flux [$\text{kg/m}^2\text{s}$] and R_s is the solid thickness or solid radius [m]. For a slab R_s is the thickness to be reached upon total shrinkage after complete drying; for a solid sphere it would be the final radius, while for a hollow sphere it would be the thickness of the solid part of the shell at a given bubble size.

Both from experimental data and from numerical calculations follows that three regimes may be distinguished, as illustrated in Fig. 11. Starting with a given initial moisture content we first have a period in which the interfacial concentration is so high that the water activity at the interface remains virtually constant : the Constant Activity Period. For a slab the flux parameter remains constant since the flux is determined by the external conditions for a given slab temperature. Inside the layer concentration profiles build up, and thus at a certain point the interfacial water concentration decreases below the point of inflection of the sorption isotherm, and so the flux will also start to decrease. Since the diffusion coefficient in that region is strongly decreasing with decreasing water concentration, the interfacial water concentration will rapidly drop to a value close to equilibrium with the drying air. If the initial flux was high, the water concentration at the non-evaporating side of the slab will still have the initial value; in that case the profiles will start to *penetrate*, until the heart is reached. We then have a Penetration Period. Once the center concentration starts to decrease we go over into the third period, the Regular Regime. The interesting observation made by Schoeber is that independent of initial conditions all drying curves, for the given layer temperature and air humidity, merge into the same Regular Regime curve. Also starting from a given moisture content at high F-values, the penetration parts of the drying curves merge into the same Penetration Period curves. When the initial flux, or rather F, is low, during the CA-period the concentration profiles will be shallow, and there is no Penetration Period, but the CA-period goes over directly into the Regular Regime.

Summarizing in the same terms as used by Schoeber we may state that :

- The Regular Regime curve is the "grandparent curve" for a given material, from which all drying curves may be derived; it does not depend on initial moisture content or initial flux.
- The Penetration Period is the "parent curve" for drying curves for a given initial moisture content at high initial F-values; the curve however does not depend on initial flux itself.
- The Constant Activity Period is determined by external conditions and initial moisture content, and thus is a kind of "child curve".

Some first rules for drying calculations with the RR-method

The basic ideas in the previous section must be refined for a full treatment. First of all the RR-curve depends on geometry; we will come back to that later. Further the RR-curve for a given geometry depends on the equilibrium value X_i of the interfacial concentration. Formally written it was found that:

$$F_{RR}(T, X, X_i) = F_{RR}(T, X, 0) - F_{RR}(T, X_i, 0) = F_{RR}^*(T, X) - F_{RR}^*(T, X_i) \quad (5-2)$$

in which $F_{RR}^*(T, X)$ is the Regular Regime curve for interfacial concentration $X_i = 0$, for a given geometry.

Thus the RR^* curve for a slab may be regarded as a material property. The dependence on the temperature is described by means of an Arrhenius equation :

$$F_{RR}^*(T, X) = F_{RR}^0(X) \exp\{-E_F(X)/RT\} \quad (5-3)$$

in which the frequency factor $F_{RR}^0(X)$ and the activation energy $E_F(X)$ both depend on the average moisture content.

For the description of a drying curve at constant layer temperature we need to know the transition points. The first one is given by the transition between PP and RR, for which the criterion is :

$$d(\ln F_{RR})/dX = 1/(X_0 - X) \quad (5-4)$$

which can be determined numerically, thus giving the transition value X_T , from which also the value of the flux parameter F_T follows. From basic considerations now follows that for the Penetration Period holds :

$$F_{PP} = (E_T F_T) / E \quad (5-5)$$

in which $E = (X_0 - X)/(X_0 - X_i)$, with X_i the interfacial moisture content. For the constant activity period the flux j_w follows from external conditions, as given by Eqs.(4-7) and (4-8), with $A_w = 1$. Intersection of the corresponding F-value with either the PP-curve or the RR-curve gives then the transition of CA to the other periods, and thus the whole drying curve is known.

For a layer the value of R_s is equal to :

$$R_s = L_0 \rho_{s,0} / d_s = L_0 / (1 + X_0 d_s / d_w) \quad (5-6)$$

in which $\rho_{s,0}$ is the initial solids concentration in $[\text{kg}/\text{m}^3]$, and d_s and d_w are the specific densities of water and dissolved solids respectively $[\text{kg}/\text{m}^3]$.

The drying time is given by :

$$t = d_s R_s \int_0^{X_0} \frac{dX}{X} \quad (5-7)$$

Short-cut estimation of aroma losses

Some 15 years ago we designed a short-cut estimation method for the loss of aroma by diffusion [16,17]. In practical spray drying also losses occur upon atomisation; with the aid of the correlation method also an estimate was made of those. Both from experiments and numerical results of the diffusion equations it appeared that in good approximation diffusional aroma loss mainly takes place during the constant activity period. Therefore a correlation method was set up in which the aroma loss is described by an effective Fourier number :

$$Fo_a = D_{a,\text{eff}} \varphi_c = D_{a,\text{eff}} t_c / R_0^2 \quad (5-8)$$

Herein $D_{a,\text{eff}}$ is an effective aroma diffusion coefficient, t_c is the length of the CA-period, and R_0 is the initial thickness of a one sided drying slab or radius of a droplet. The relation between the fractional aroma retention and the Fourier number may be approximated by :

$$AR = \sum_{n=0}^{\infty} A_n \exp \left[- \beta_n^2 Fo_a \right]$$

$$A_n = \frac{8}{\pi^2} \frac{1}{(2n+1)^2} \quad \beta_n = 2 / A_n \quad \text{slab} \quad (5-9)$$

$$A_n = \frac{6}{\pi^2} \frac{1}{n^2} \quad \beta_n = 6 / A_n \quad \text{solid sphere}$$

or by the well-known short-time and long-time approximations :

$$\left. \begin{aligned} AR &= 1 - 2 (Fo_a / \pi)^{1/2} & Fo_a < 0.2 \\ AR &= \frac{8}{\pi^2} \exp \left[-\pi^2 Fo_a / 4 \right] & Fo_a > 0.2 \end{aligned} \right\} \text{slab} \quad (5-10a)$$

$$\left. \begin{aligned} AR &= 1 - 6 (Fo_a / \pi)^{1/2} & Fo_a < 0.022 \\ AR &= \frac{6}{\pi^2} \exp \left[-\pi^2 Fo_a \right] & Fo_a > 0.022 \end{aligned} \right\} \text{solid sphere} \quad (5-10b)$$

The length of the CA-period was correlated to the initial flux and other parameters by means of the following :

$$\varphi_c = \frac{B}{\varepsilon} \quad \text{slow drying} \quad (5-11a)$$

$$\varphi_c = \frac{F_w}{\varepsilon^2} \quad \text{fast drying} \quad (5-11b)$$

Herein ε is a somewhat different kind of flux parameter :

$$\varepsilon = j_{w0} R_0 / d_w \quad (5-12)$$

with j_{w0} the initial water flux. The value of B can be derived for totally flat water concentration profiles :

$$B = \frac{(X_0 - X_c) d_s / d_w}{(1 + X_0 d_s / d_w)} \quad \text{slab} \quad (5-13)$$

$$B = \frac{1}{2} \left[1 - \left[\frac{1 + X_c d_s / d_w}{1 + X_0 d_s / d_w} \right]^{2\beta} \right] \quad \text{solid sphere} \quad (5-14)$$

Herein X_0 is again the initial water content on dry base, and X_c is the *critical interfacial moisture content*; for X_c the value may be taken at which according to the sorption isotherm $A_w = 0.90$. While for this regime of extremely slow drying only X_c appears as material property, for the case of fast drying the diffusion properties enter,

as dependent on initial moisture content and sample temperature. This could be well correlated by :

$$F_w = F_w^0 \exp \left[- \frac{E_w}{RT} + f_{ww} \rho_{w0} / d_w \right] \quad (5-15)$$

For aqueous maltodextrin solutions the following values were found :

$$X_c = 0.429 \quad F_w^0 = 1.35 \cdot 10^{-9} \text{ m}^2/\text{s} \quad E_w/R = 2764 \text{ K}^{-1} \quad f_{ww} = 8.781$$

The question whether to consider a specific situation as slow or fast drying is easily solved by calculating both values of ϕ_c according to Eq.(5-11) : the lowest value of ϕ_c should be taken.

The effective aroma diffusion coefficient may be correlated by :

$$D_{a,\text{eff}} = D_a^0 \exp \left[- \frac{E_a}{RT} + f_{aw} \rho_{w0} / d_w \right] \quad (5-16)$$

In Fig. 12 a plot is given of $D_{a,\text{eff}}$ vs. the solids concentration for the three components for a temperature of 50°C ; as could be expected the effective diffusion coefficient decreases with increasing solids concentration and with increasing molecular mass of the aroma component.

In recent years more accurate methods have been developed to predict t_c [15]; also the loss of aroma has again been subject of more accurate measurement, as presented by by Coumans et al [10]. Thus in future a further refinement of this correlation method may be made. Still needed, but within reach, is the correlation for hollow spherical particles.

One of the interesting aspects of this short-cut method is that the basic data can again be determined from simple lab experiments in which layers of the material are dried. Thus the tedious determination by differential measurements, as well as the numerical computations, can be avoided.

thermal degradation

Again the use of short-cut methods would be welcome. As suggested in 1977, one very simple approximation would be to approximate the degradation rate by that for the *average*

Fig.12. Effective aroma diffusion coefficients for n - alcohols in aqueous maltodextrin solutions

moisture content, which is readily available from short-cut calculations [18]. Although this will give some deviations it will still be a handy tool in the predictions of trends.

SPRAY DRYING OF LIQUID FOODS

Approximation for spray drying of hollow particles

For spray drying we have in the majority of processes *hollow spherical particles*. From simulations it followed that for thin shells we can successfully approximate the drying curve by that of a drying layer, provided that we use the following definitions :

$$R_s = (R_{out} - R_{in}) / (1 + X d_s / d_w) \quad (6-1)$$

As of course the temperature of a drying particle is variable we will see a change in the inner and outer radius. For foods a good approximation is to assume a zero pressure

difference between inside and outside. Thus follows at any time :

$$(P_w + P_{air})_{in} = P_{tot} \quad (6-2)$$

$$\text{with } P_{w,in} \approx A_w(X) \cdot P_{w,s} \quad (6-3)$$

The inner radius at any time is then given by the conservation of included air :

$$R_{in}^3 P_{air} = (R_{in}^3 P_{air})_0 \quad (6-4)$$

Now during the CA-period the temperature remains constant, at wet-bulb temperature, and consequently the inner radius remains constant. Due to water loss the outer surface shrinks, and thus R_s will also change. Since for the gas-phase Sherwood-number approximately holds :

$$Sh_c \approx 2 \quad (6-5)$$

the mass transfer coefficient in the gas phase varies roughly proportional to the outer radius :

$$k_g \approx D_{wg}/R_{out} \quad (6-6)$$

and for not too high evaporation rates :

$$k_{eff} \approx k_g \quad (6-7)$$

Thus the flux for the CA-period follows from Eq.(4-7), and R_s is calculated according to the above.

For the variable temperature it has been demonstrated by the mentioned authors that the RR-curves and PP-curves may be found by stepping over between the curves for constant temperatures. Thus we may divide the process into the following steps:

1 Find the wet-bulb temperature. Determine numerically the intersection point between either the PP or the RR and the CA-curve at this temperature. Find $F = F_{ca}(X)$ by calculation of R_s and j_w over a number of intervals.

2 Below the average critical moisture content take a small step in X . Calculate a piece of the drying curve in the prevailing regime, PP or RR, at the interfacial water concentration corresponding to the temperature. Apply the mass and enthalpy balances :

$$\frac{dm_w}{dt} = \frac{d}{dt} \left[m_s X + m_g X' \right] = -j_{w,i} S = -F S / R_s \quad (6-8)$$

$$\frac{dJ}{dt} = \frac{d}{dt} \left[m_s j + m_g i \right] = (F_H - F) S \Delta h_v / R_s \quad (6-10)$$

$$F_H = \alpha_{eff} (T_b - T) R_s / \Delta h_v \quad (6-11)$$

in which X' is the absolute humidity inside the gas bubble, Δh_v is the heat of evaporation, and S is again the outer surface area. J stands for the total enthalpy content.

From these balances follow the changes in moisture content X of the shell and in temperature :

$$\frac{dm_w}{dt} = \left[\frac{dX_w}{dt} \right] \left[\frac{\partial m_w}{\partial X_w} \right]_T + \left[\frac{dT}{dt} \right] \left[\frac{\partial m_w}{\partial T} \right]_{X_w} \quad (6-11)$$

$$\frac{dJ}{dt} = \left[\frac{dX_w}{dt} \right] \left[\frac{\partial J}{\partial X_w} \right]_T + \left[\frac{dT}{dt} \right] \left[\frac{\partial J}{\partial T} \right]_{X_w} \quad (6-12)$$

The partial derivatives follow from numerical differentiation of relations for m_w and J , and so the changes in X and T can be solved.

Thus a table of values of F , R_s and R_{out} can be made up over the whole concentration range in X . The drying time is then found by integration :

$$t = \frac{m_s}{4\pi} \int_X^{X_0} \frac{R_s dX}{F R_{out}^2} \quad (6-13)$$

aroma retention

In Fig. 13 the aroma retention for the three alcohols has been plotted according to the correlation method for a typical situation in our pilot spray drier, as reported by

Fig.13. Predicted aroma retention in spray drying model aroma components compared with experiments

Rulkens and Thijssen [19]. From the mass balance over the drier the air humidity was calculated for the range in feed concentrations, and with the above the aroma retention was estimated for solid spheres. As can be seen the experimental data are roughly at 70% of the predicted ones with this method. This may partly be due to losses on atomization, partly because in reality the particles were not solid but hollow, which probably would lead to lower theoretical retentions as well. We see however that the trends found experimentally are reflected also in the predictions. The increase in retention with increasing solids concentration is caused by the lower aroma diffusion coefficient and the shorter length of the CA-period, as indicated in Fig. 14.

thermal degradation

This is illustrated in Fig. 15, in which we calculated the drying history of skim milk hollow particles in air of 70 °C and a humidity of 0.0518 kg/kg, with an initial gas volume of 20 %. Simultaneously we integrated the reaction rate at the average

Fig.14. Reduced length of constant-rate period in spray drying of aqueous maltodextrin solutions

Fig.15. Rest activity after spray drying skim-milk particles, calculated with a model degradation reaction (fig. 10) on the basis of the average water content

concentration and prevailing particle temperature. We see a very large influence of the initial droplet diameter on the estimated final rest activity, which is caused by the longer drying and thus reaction times for such particles, the times being proportional to a power of R_0 between 1 and 2, depending on the Biot number. This effect is also found in more detailed calculations.

Simple approximation of particle motion in spray drying

Although at present considerable effort is taken to model the motion of both particles and air in spray driers by means of Computational Fluid Dynamics, this has as yet not led to user-friendly applications. For the purpose of estimation a relatively simple approximation is found in the single-particle motion [8]. This model lends itself well for both uniform air flow and for swirling motion, and since it is of the same kind of numerical complexity as the short-cut methods for drying particles it can easily be integrated with those.

For the equation of motion we may write :

$$m \frac{du}{dt} = (m - m')g - C_w A_{cs}^{1/2} \rho_l w |w| \quad (6-14)$$

Herein m is the particle mass, m' the mass of displaced air, u the velocity vector of the particle with respect to stationary coordinates, g the gravity acceleration vector, C_w the drag coefficient, A_{cs} the cross-sectional area and ρ_l the density of air. The relative velocity of the particle with respect to that of the air v is denoted by $w = u - v$.

The drag coefficient C_w depends on the Reynolds number; a continuous calculation can be made by means of the correlation of Kaskas [20] :

$$Re \leq 2.10^5 \quad C_w = \frac{24}{Re} + \frac{4}{Re^{0.5}} + 0.40 \quad (6-15)$$

in which $Re = \rho_l |w| d / \mu_l$.

The absolute value of the velocity difference vector, $|w|$, is given by :

$$|w| = [w_x^2 + w_y^2 + w_z^2]^{1/2} \quad (6-16)$$

Fig.16. Horizontal and vertical trajectories calculated as single particle behaviour, initial velocity 200 m/s at 60 deg angle with downward vertical

The trajectories follow by simple integration of the velocity components over time :

$$S = \int_0^t u \, dt \quad (6-17)$$

As an example in Fig.16 the horizontal and vertical trajectories of hollow droplets with an internal gas volume of 20% have been calculated for the case of an initial velocity of 200 m/s under an angle of 60 degrees with the downward vertical. The air is assumed to have a uniform downward velocity of .532 m/s. It is clear that the smaller droplets have a much shorter deceleration path than the larger ones.

In reality the trajectories will be much more complicated; one may expect that the deceleration path is longer because of the air flow induced by the atomisation itself. However for first estimates and for comparison of situations this approximation has proved to be a useful tool.

Fig.17. Number and mass based frequency distribution of droplet sizes for calculation example

Integration example

As treated in the foregoing, there are a number of more or less complex methods to estimate drying rate, aroma loss, thermal degradation and trajectories for single particles. In order to illustrate the integration of the methods we made an example calculation of a drier with a diameter of 9 m and a height of 16 m, in which 5000 kg/hr of skim milk was to be made with a moisture content of 7 %. We assumed an initial droplet size distribution as given by Masters [21] as an example. This number based distribution was calculated into a mass based distribution, as given in Fig. 17. Now assuming disk atomisation with an initial velocity of 200 m/s, the vertical trajectories were calculated vs. time of flight, as given in Fig. 18. From this graph the residence times of the particles could be estimated for the specified height. Also the drying times for the specified air conditions were calculated as a function of moisture content, as presented in Figs. 19a and 19b. So for each particle class the moisture content followed from its

Fig.18. Vertical trajectories calculated as single particle behaviour, initial velocity 200 m/s at 0 deg angle with downward vertical

residence time. We also assumed for the case of this example that some thermal degradation reaction would occur, corresponding to that treated in the previous chapter. Both the water content and the rest activity were then integrated over the whole distribution. The individual class results, together with the mass averaged values are shown in Fig. 20. It is clear that in this type of distribution the higher moisture content in the larger classes leads to a moisture content well above the equilibrium value. Due to the zero rest activity in the larger particles also the average activity is very low.

BATCH FLUIDISED DRYING OF BIOPRODUCTS

Effective heat and mass transfer coefficients

The basic well-known model is that of a bed in which the particles are ideally mixed, through which the air flows in plug flow, as has been used by Zimmermann [22] and

Fig.19. Drying time vs. moisture content for hollow skim milk droplets , calculated by means of the short-cut method. Air conditions : 70 deg. C, $H = 0.052$ kg/kg. Initial vacuole volume 30 %.

(a) larger particles (b) smaller particles

Fig.20. Distribution of final moisture content and rest activity over the droplet size distribution, together with mass averaged values

Strumillo [23], and illustrated in Fig.21. In a very thorough study Lievens et al [24,25,26] recently described drying and inactivation kinetics of pellets of a mixture of *Lactobacillus Plantarum* and starch. It was thought of some interest also for educational purposes to translate their findings from laboratory experiments into considerations for large scale design. For the sake of the example no attention was paid here to the question of drying during filling of the bed, as would have to be taken into account in practice.

Differential balances over the drying air

Following the assumption of an ideally mixed particle bed we assume that we may approximate the surface water concentration and the particle temperature by average values over the whole bed. From a differential height element we have the balances over the air :

$$V \frac{dH}{dz} = k_{g,loc} \rho_f (H_i - H) a_p (1 - \epsilon_b) A_b \quad (7-1)$$

Fig.21. Diagram of batch fluidized bed drier, with air flowing through in plug flow and solids ideally mixed

$$V \frac{d i}{d z} = \left[\alpha_{loc} (T - T_{air,z}) + k_{g,loc} \rho_f (H_i - H) i_v \right] a_p (1 - \epsilon_b) A_b \quad (7-2)$$

in which a_p is the specific area of the particles [m^2/m^3], ρ_f is the film density of the air [kg/m^3], ϵ_b and A_b are the bed porosity and diameter respectively. The coefficients $k_{g,loc}$ and α_{loc} are the local mass and heat transfer coefficients [m/s] and [kW/m^2s] respectively. The enthalpy of the created water vapour is denoted by i_v . The particle temperature is denoted by T .

By using the psychrometric relationship :

$$i = (c_a + c_v H) T_{air} + H \Delta h_v \quad (7-3)$$

in which c_a and c_v are the specific heats of air and vapour respectively, and some approximations we may derive an approximation for the air temperature. Introducing the number of thermal transfer units N_{th} and the number of mass transfer units N_{tm} according to the following definitions :

$$N_{th} = \frac{\alpha_{loc} a_p M_p}{\rho_p V c_{a,av}} = \frac{\alpha_{loc} A_{pt}}{V c_{a,av}} \quad N_{tm} = \frac{k_{loc} a_p \rho_f M_p}{\rho_p V} = \frac{k_{loc} \rho_f A_{pt}}{V} \quad (7-4)$$

and the dimensionless bed height $\xi = z / L$

we may derive for the humidity and temperature of the drying air :

$$H - H_i = (H - H_{in}) \exp \left[- N_{tm} \xi \right] \quad (7-5)$$

$$T_{air} - T = (T_{air,in} - T) \exp \left[- N_{th} \xi \right] \quad (7-6)$$

In the above A_{pt} is the total exchange area in the bed.

Effective heat and mass transfer coefficients around the particle

From considerations of a particle which is exposed to random surrounding conditions it follows that effectively the mass and heat transfer rate may be approximated by that obtained by taking the height averaged driving forces over the bed. We may write the average fluxes by relating the driving force to the bed entrance conditions :

$$j_q = \alpha_{eff} (T_{air,in} - T) \quad (7-7)$$

$$j_w = k_{g,eff} \rho_f (H_i - H_{in}) \quad (7-8)$$

Applying this we find for the effective transfer coefficients :

$$\alpha_{\text{eff}} = \alpha_{\text{loc}} \frac{1}{N_{\text{th}}} \left[1 - \exp(-N_{\text{th}}) \right] \quad (7-9)$$

$$k_{\text{g,eff}} = k_{\text{g,loc}} \frac{1}{N_{\text{tm}}} \left[1 - \exp(-N_{\text{tm}}) \right] \quad (7-10)$$

For the local heat transfer coefficient we use the relation of Nelson and Galloway [27] :

$$\text{Nu}_{\text{loc}} = \frac{\alpha_{\text{loc}} d_p}{\lambda_{\text{air}}} = \frac{2\varphi + \left\{ \frac{2\varphi^2 (1-\epsilon)^{1/3}}{[1 - (1-\epsilon)^{1/3}]^2} - 2 \right\}}{\frac{\varphi}{1 - (1-\epsilon)^{1/3}} - \tanh \varphi} \quad (7-11)$$

$$\text{in which } \varphi = 0.3 \left[\frac{1}{(1-\epsilon)^{1/3}} - 1 \right] \text{Re}^{1/2} \text{Pr}^{1/3} \quad (7-12)$$

The local mass transfer coefficient follows from the Chilton - Colburn analogy :

$$k_{\text{g,loc}} = \frac{\alpha_{\text{loc}}}{\rho_f c_{\text{pf}}} (\text{Le})^{-0.33} \quad (7-13)$$

in which Le is the Lewis number $\text{Le} = \text{Sc}/\text{Pr}$.

In the foregoing d_p is the effective particle diameter. For cylindrical pellets this is equal to :

$$d_p = \frac{3}{2} \frac{\beta}{(\beta + 0.5)} d_c \quad (7-14)$$

with $\beta = d_c / L_c$, the ratio of the cylinder diameter and length.

Fluidisation characteristics

For the minimum fluidisation velocity we use the relation of Davidson and Harrison [28] :

$$\text{Re}_{\text{mf}} = 25.7 \left[\left(1 + 5.53 \times 10^{-5} \text{Ga} \right)^{1/2} - 1 \right] \quad (7-15)$$

with $\text{Ga} = \rho_f \Delta \rho g d_p^3 / \mu_f^2$, the Galilei number.

For the relation between bed porosity and actual air velocity we approximated the results reported by Keey [29] :

$$\varepsilon = 1 - 0.65 \exp [-0.27 u / u_{mf}] \quad (7-16)$$

Internal mass transfer and equilibrium

As reported by Lievense the mass transfer can well be described by means of an effective diffusion coefficient, dependent on water concentration and temperature :

$$D = 1.5 \times 10^{-10} X^{1.1066} \exp \left[-3404.9 \left(\frac{1}{T_{abs}} - \frac{1}{303.15} \right) \right] \quad (7-17)$$

From estimations of the thermal Biot-number follows that the temperature gradients inside the particle can be neglected.

For the water vapour sorption isotherm he reports the G.A.B. isotherm :

$$\frac{X}{X_m} = \frac{C k a_w}{(1 - k a_w)(1 - k a_w + C k a_w)}$$

with $X_m = 0.0794 \text{ kg H}_2\text{O} / \text{kg ds}$, $C = 28.0$, $k = 0.859$

Simulation results

Lievense et al. solved the diffusion equation (4-3) for infinite cylindrical particles by means of the power-law short-cut calculation method of Liou and Coumans. They assumed a very short constant activity period, and so immediately started the calculation for the Penetration Period and subsequently the Regular Regime. They assumed virtually immediate temperature equilibrium between the 1 mm particles and air. This was justified in their situation, because *in the lab drier they used a very small amount of product (10 g.) and a large airflow (50 kg/hr)*. Consequently they had a low nr. of transfer units and thus relatively high transfer coefficients : $N_{th} = 0.93$, $N_{tm} = 0.99$, leading to $\alpha_{eff} = 0.24 \text{ kW/m}^2 \text{ } ^\circ\text{C}$, and $k_{eff} = 0.22 \text{ m/s}$.

Fig.22. Moisture content and temperature of granules of *L. plantarum* during laboratory fluidized bed drying, for different bed loads

Since their primary aim with the lab experiments was to obtain data for the drying kinetics, it was of interest to see to what extent a drying curve would be influenced by the bed load. We solved the diffusion equation by means of a modified Crank-Nicholson scheme, together with the enthalpy balance for the particles. In Fig. 22 the drying curves are given for the lab drier, for loads of 10 and of 100 g. It is clear that for the small load the temperature rise is virtually immediate, while for the larger load it takes considerably longer. The drying rate for the larger load is somewhat smaller. From this can be seen that *the theoretical model is a very useful tool in deciding upon experimental design !*

It is clear that in practice one wants to make optimal use of the drying air. Thus the bed loads will be much higher than in the lab. A simulation was therefore also done with a bed load of 1000 kg, for a bed with a diameter of 3 m, and an air flow rate of 50000 kg/hr. Again the air temperature for the example was taken to be 30 °C. In Fig. 23 we see that the drying curve is dramatically different in shape from that in the lab drier. The temperature first drops and rises only very slowly. This is caused by the much

Fig.23. Predicted moisture content and temperature during fluidized bed drying on large scale for a bed load of 1000 kg

Fig.24. Water concentration profiles inside drying particles of *L. planetarium* as calculated by solving the diffusion equation. Left figure : laboratory drier with 10 g. load, right figure : large scale fluid bed, 1000 kg.

higher number of transfer units : $N_{th} = 50$, $N_{tm} = 53$, leading to very low effective heat and mass transfer coefficients of $0.004 \text{ kW/m}^2 \text{ } ^\circ\text{C}$ and 0.0035 m/s respectively.

To illustrate the differences further, we have plotted in Fig. 24 the internal moisture concentration profiles. For the lab situation we see very steep concentration profiles, for the full scale drier they are much more shallow. Clearly the mass transfer limitation has shifted from fully within the particle to a much more pronounced influence of the external resistance. To return to the discussion in the beginning of this paper : for the lab drier the modified Bi-number can be estimated to be 300, while for the full scale it is about 4.

Since inactivation of microorganisms locally inside the pellets is determined by the local drying and temperature history, the overall effect may be expected to be considerably different for lab and full scale drying.

CONCLUDING REMARKS

In the foregoing we have implicitly adressed modelling aspects on three levels : *physical, mathematical and simulation models*. In conclusion the relevance of these types of modelling for scale-up will be summarized in the following.

Physical modelling, setting up an explicit picture of the (sub) processes going on in the drying processes in the lab and full scale, is probably the most important step of all. It provides a framework for communication between various parties involved in the field of scale-up, and urges them to formulate insight into the process. This in itself, combined with some estimations of the magnitude of effects in different situations, may already improve scale-up practice and drying operations.

Mathematical modelling, the translation of the various physical ideas into quantifiable rules, is the key between reasoning and robust predictions. It requires the application of sound engineering principles, and defines what quantities and relationships should be looked for either in literature or from experiments. It also gives insight into the combinations of process variables, and in some cases even without any calculations at all similarity analysis may lead to scale rules. From the diffusion equations with boundary conditions it follows for instance that one would expect only a very small influence of droplet size on aroma retention, but a dramatic effect on thermal degradation [8,18].

In *simulation models* the foregoing modelling techniques are put to work. With the modern 386/387 PC's it has become possible to attack a lot of the complex non-linearities in a very rigorous way in a few minutes of calculation time. Combining the calculation models with the very user-friendly graphic programs has opened the possibility of making various phenomena visible, and study the effect of scale and of process variables, as has been shown in the examples. As has been done in the chemical industry for decades, also in drying operations first simulations can be performed before buying or designing equipment. Although good physical data are needed for a proper description, already a lot can be learned from simulations of the drying behaviour of analogue materials.

An essential element is that PC - programs are easily transferred. Although this may incidentally lead to use without full understanding, if combined with a proper description it has the great advantage of opening up the possibilities of simulation for a wider public. Thus the incorporation of complex phenomena in scaling up can be more and more implemented in daily practice.

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APPENDIX. REGULAR REGIME AND DIFFUSION DATA

Regular Regime data for skim milk and maltodextrin solutions have been fitted to the general form

$$\ln F_{RR}^* = a - b \left[\frac{273.15}{T} \right] - \left[\frac{X}{c + X} \right] \left\{ d - e \left[\frac{273.15}{T} \right] \right\}$$

with T the absolute temperature, and X the moisture content on dry basis.

It is advised not to exceed the range of $T > 350$ K, especially at low moisture concentrations.

Also for the molecular diffusion coefficient of water a similar fit was made :

$$\ln D = a_1 - b_1 \left[\frac{273.15}{T} \right] - \left[\frac{X}{c_1 + X} \right] \left\{ d_1 - e_1 \left[\frac{273.15}{T} \right] \right\}$$

	a	b	c	d	e
skim milk, sphere	10.49	44.81	0.12	17.23	37.46
skim milk, slab	11.75	45.84	0.12	20.04	38.70
md20, sphere	50.07	84.77	0.18	61.66	82.72
md20, slab	52.44	87.47	0.18	66.27	86.24
$D_{\text{skim milk}}$	6.49	42.99	0.12	22.86	37.64
D_{md20}	39.32	77.42	0.18	63.38	80.45

Effective aroma diffusion coefficients used in correlation method

For n-alcohols in aqueous maltodextrin solutions E_a/R was found to be equal to 5000 K^{-1} ; the values of D_a^0 and f_{aw} were found to be :

component	D_a^0 (m ² /s)	f_{aw}
methanol	$1.86 \cdot 10^{-4}$	5.41
n-propanol	$3.84 \cdot 10^{-5}$	6.65
n-pentanol	$2.26 \cdot 10^{-5}$	7.04