DRYING, GROWTH TOWARDS A UNIT OPERATION

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ABSTRACT
Within the field of separations drying is one of the unit operations with a very wide variety of materials, equipment type, capacity, product requirements and added value. The scope and depth of published papers about the subject shows a similar variety. At the present time the systematic approach to design and operation, as appears to exist for other unit operations, for drying is still largely incomplete. In the paper some aspects of unification are considered. After some thoughts about types of drying research, examples are given from thermodynamics, transport phenomena and mathematics to show perspectives of using common concepts and theory towards fruitful integration of knowledge from other areas into drying knowledge.

INTRODUCTION
One of my personal incentives for doing engineering science can be summarized by the motto “Today’s Science for Tomorrow’s Tools”; in consulting it is “And of course, Yesterday’s Science for Today’s Tools”. Let me state clearly at the beginning that this is going to be a very personal contribution, the good things inspired by the excellent work of a lot of other scientists and engineers, the bad things due to my own limitations. Within the field of separations drying is one of the unit operations with a very wide variety of materials, equipment type, capacity, product requirements and added value. The scope and depth of published papers about the subject shows a similar variety. At the present time the systematic approach to design and operation, as appears to exist for other unit operations, for drying is still largely incomplete. Many workers both in academia and industry do not appear to know about or use the excellent work done by top scientists in their area, and about the heritage of classic work. If we compare the state of the drying field to that of other mature unit operations, like distillation and extraction, we can see that the difference lies in the term “unit”, or rather unity. More in detail we can
have a look at unity in physical concepts, theory and methodology. In the following I will first muse a little bit over our differences within the drying community, and possible reasons for them. The main part of the paper will be dedicated to a selection of topics where I will discuss unifying concepts within the drying area, as well as the possibility of increasing shared knowledge and methods with those from other areas.

In the IDS conference proceedings and the Drying Technology journal we have a very large mix of contributions, from scientific to very practical. Following my first motto it may be interesting to consider the area of scientific research a bit more. As in many countries, also in the Netherlands we have discussions at governmental level about the quality and quantity of research, and many attempts to provide information in a form suitable for politicians. In one of the reports a nice diagram was presented of Berkhout (1998), presented in Figure 1, which we can also try to apply to drying research and development. For the people from industry, such as equipment manufacturers, applied science institutes and end-users, their mission will be clearly in the direction of societal relevance, and so in the scheme one would expect applied scientific research. Looking at the methodology, their aim would be at integrated results, such as a novel dryer or dryer configuration, a better control system, an improved or novel product. Thus one could see that the activities of these workers would be mainly focused in area 3 of the matrix. As indicated, such work would have and need interaction with people in areas 2 and 4. If we look at area 2, so the combination of discipline oriented methodology, and societal relevance as a mission, we can see very clearly the control researchers as examples. A number of them is involved in developing and implementing e.g. fuzzy logic algorithms for various dryers, or optimizing drying schedules. Also instrumentation developers work for a part in this areas making novel sensors. As examples of activities in sector 4 we may see the CFD modeling of spray dryers, many-particle models in fluidization, and modeling of mass, heat and momentum fields in deforming materials. An example in section 1 would be the volume-averaging theory of Whitaker et al. (1977, 1998). An interesting party game could be to ask other people to put your research into the matrix, and compare with your own positioning, and have a discussion about this. (In our country we have such parties under the name of management courses, they are very nice but tend to be rather costly). Anyway, especially if you are working in academia or scientific institutes, some reflection on what your drive is and your way of attacking subjects, can always be helpful. Going through the conference proceedings of the present and some past IDsa, it is my impression that in the first place there are still bridges lacking between areas 3 and 4: many applied developers do not use the results of people involved in process modeling. The same holds for areas 2 and 3; here I sometimes have the impression that the discipline-orientated researchers do not pay enough attention to the reality of the systems they try to model or control. In area 1, pure scientific research, we have a significant number of contributions, some of which will find the way to numbers 4. In general my personal impression is that the bridging between 1 and 4 is a bit stronger than the others, but still can be strengthened. Next to regarding the areas as positions for present research activities, they may also be

Figure 1. Schematic view of positioning research activities
viewed as reservoirs of existing knowledge, either scientific or experience. One of the striking things in drying is the very large range in awareness of what is in these reservoirs; some authors build on the finest scientific traditions and add to it, others are totally unaware.

The diagram shown is 2-dimensional, and as a third dimension you could add quality. Again, the range is very wide; we see papers of very high and of very low quality, and everything in between. Realizing that every manuscript represents a more or less substantial amount of man-months to years of work, one can conclude that next to very rewarding efforts also a lot of work will neither give the authors any further satisfaction nor promote drying science and technology.

**Funding**

All research and development in any area has to be funded, and so we may speculate a little bit about the decisions made with regard to this in the drying area. In different countries there is a remarkable difference in providing funds for basic research in drying; in the Netherlands it is definitely not a hot item for the scientific agencies when compared to catalysis, macro-organic chemistry or nano-technology. Viewing the proceedings of this conference it is clear from the acknowledgements that e.g. in Brasil there is a very large support from governmental and state agencies, and also that most of the work is done in the area of foods and agricultural products. In the very large AIChE annual meetings drying does not or only marginally occur in the conference programs, reflecting the very low number of university research groups active in the area. In Scandinavian countries, New Zealand and Canada a lot of work is done in drying of wood and derived products such as boards and paper. Without attempting to make a detailed evaluation we may conclude that for large application areas there is a strong relation between the subjects and attention for drying research and local economic factors. In a minority of situations there is felt a natural national or regional need from both industry and government that applied development has to be supported by basic research, and a dedicated institution has been founded, mostly combining both and liaising to university groups. Drying is known to be very energy-intensive; in Western Europe the operation uses around 11% of total industrial energy consumption. In order to decrease CO₂-emissions as laid down in the Kyoto-agreement therefore also drying is one of the fields to work at. Funding for process improvement in this area by drying research has not reached any substantial international levels. In a minority of cases one can be lucky to find industrial sponsoring; in a fraction of those one can even obtain it for basic research. Still the targets in present industry are directed more to new and wider ranges of products, higher quality and throughput. This could accord well with good research and development, however often the industrial time-scale is one order of magnitude smaller than that needed for knowledge development.

So for most of us we can only do our drying research if at least we sell our projects under the flag of societal relevance, and try to influence our funding agencies to set up programs as we like them best, but also marketable to their political superiors.

**The quality and quantity factors**

The quality of research results depends on the people doing the research, and on the circumstances in which they work. In academia as well as in institutes it is the responsibility of the leaders to provide challenges as well as guidance to their students and researchers. For every project also a knowledge base is needed, which the student or new researcher has to master. The existence of the large reservoirs of knowledge mentioned above can be very helpful when guided by the experienced, however the size may make it a mer à boire for the uninitiated. Structuring our knowledge into a common framework would be of tremendous value.

There are several things one can worry about in drying science. One is that it may be difficult to keep continuity at the top of the field. This however worries me less than the fact that in many papers at the bottom of the field the authors show only a marginal grasp of writing scientific papers, and/or of doing scientific work. Next to the “closed-loop research” danger as signaled by Mujumdar (1998) for work
which is only done and read by academics, there is also a considerable “autarky” danger. With this I mean that groups disregard or are unaware of scientific basics both in drying and underlying physics, and even mathematics, and in part try to re-invent wheels and even create their own pseudo-science. When such things happen, we may conclude that the scientific community has not been able yet to provide sufficiently clear standards and to form a jointly accepted framework, and so this is one of the challenges for the future.

For quite some time it has been very hard for a number of our colleagues to simply have access to all kinds of literature, thus causing relative isolation. I sincerely hope that with the possibilities of Internet on one hand and the enthusiastic projects on databases which are reported also in the present IDS, that these limitations will be taken away in the time to come. Then there is the question what to study, and what to put into the databases. This will ask for a filtering process, which will need a lot of attention and time. In a field which would seemingly be much more coherent, molecular modeling, there is an AIChE scientist taskforce setting up an Internet handbook, with on-line computation examples (http://w3press.utk.edu). From the start in 1995 until now only a limited amount of material has become available, but of high quality, and so we may conclude that for our field this will also be a long-term effort.

It is of importance to obtain structure in the outlet of published information. A considerable number of contributions is concerned with the determination or estimation of physical properties, sometimes implicitly. Analogously to the J. Chem. Eng. Data, we might dedicate a part of Drying Technology and of our future databases to collection of such data. As follows from critical reading it is definitely necessary that standards are set up with respect to the experimental procedures and the evaluation of physical properties.

In the following I have chosen, arbitrarily, some areas in which it is maybe possible to make some steps towards unifying our concepts. Mostly I will limit myself to the systems in which water is removed, often by means of a convective gas flow.

**STATIC PHYSICAL PROPERTIES**

*Psychrometry*

One of the areas in which most workers now employ a common base is that of psychrometry. A very large part of drying practice is concerned with water removal by air, and the classic approach for humidity and thermal properties seems to have penetrated over a large area. Still occasionally there is confusion between the relative humidity \( RH = 100 \frac{P_w}{P_{w,sat}} \approx 100 a_w \) and the relative absolute humidity \( 100 \frac{H}{H_{sat}} \). The basics are well described in books like Krischer and Kast (1978) and Keey (1972). A bit more complicated is the treatment of combustion gases, because water content and temperature are correlated, and because often the temperature range is so large that no longer constant specific heats can be assumed. In both areas several computer programs are available; a good overview is given by Menshutina and Kudra (2000), including non-aqueous systems. In making balances over equipment the convention of expressing quantities on dry gas and dry solids basis has found wide acceptance. One of the factors sometimes not taken into account is the total pressure which in general will be different from 1 bar, depending on location in equipment and geographic circumstances. Next to numeric values calculated in computer programs, graphical representations of gas properties and drying trajectories may add to the insight into the drying process.
Sorption isotherms, use in drying considerations

As in many other unit operations one needs physical data about phase equilibrium between drying material and surroundings, as represented by the water vapour sorption isotherm. The following may sometimes be quite elementary; however inspection of the manuscripts shows that some contributors do not have any idea about the existence of these isotherms. An example is presented in Figure 2 for aqueous maltodextrin solution, in which the equilibrium water activity is given here against the water content, for three temperatures. This illustrates the quite common features as are found in many materials. At high moisture content the water activity is very close to 1, and also the temperature effect becomes very small. At lower moisture content the water activity shows a rather steep dependence on water content and the temperature influence is clearly there. The first significance of the sorption isotherm is that it presents the drying curve of a material being dried under equilibrium conditions. This may be approached when drying very small particles in large beds with small amount of air. When there are no internal moisture gradients, the actual water activity in the air will correspond to the equilibrium value as given by the isotherm. The next significant thing to be read from it is the ultimate moisture content attainable in a drying process. At the product outlet there is a certain relative humidity, or water activity, and the product in general cannot be drier than the moisture content given by the isotherm. (One can construct schemes in which this would be the case, however mostly they would present badly constructed dryers). The third aspect to be seen is related to the often-called "first period of drying". For most materials that we dry there will be internal moisture concentration gradients, and so during the process the moisture content at the surface will decrease. As can be seen in the figure the equilibrium water concentration will also decrease. For the water flux at the surface we have:

\[ j_w^i = k_f (\rho_{wg}^i - \rho_{wg}^b) = k_f \frac{M_w}{R} (a_w^i - a_w^b) \frac{P_{w,sat}(T^i)}{T^i} - a_w^b \frac{P_{w,sat}(T^b)}{T^b} \]  

Let us assume for now that we have from the beginning thermal balance between water evaporation rate and heat transfer to the material. Then the temperature at the interface \( T^i \) will be constant, and so will the saturated vapour pressure \( P_{w,sat}(T^i) \). In Figure 3 some results are given of a simulation of a non-hollow droplet of skim milk in a well-mixed spraydryer. Here the surface water activity \( a_w^i \), the average water fraction \( <\omega_w> \), and the surface water fraction \( \omega_w^i \) are given as function of the drying time. We start with a high moisture content, 70% water, and the interfacial water activity will be very close to 1, and while the surface concentration decreases it still remains close to 1, until the surface water concentration decreases below a value where the isotherm goes “into the bend”, in the example here at about 10% water. The drying rate for isotropic drying is given by:

\[ \frac{dW_w}{dt} = W_s \frac{dX_w}{dt} = -A_p j_w^i \]
For shrinking materials it may well be that both the sample surface area \( A_p \) and the mass transfer coefficient \( k_f \) will change in different ways, and so neither the drying flux nor the drying rate will be constant. In Figure 4 it is very clear that also in the first part of the drying curve the drying rate is variable due to the shrinkage; the drying flux looks more constant but still is a little variable. If you are drying a slab with crossflow of air, or nondeformable samples, the initial rate and flux will remain constant, still provided the temperature is at the equilibrium value (often close to wet-bulb temperature) from the start. In general the initial product temperature differs from the wet-bulb temperature, and so you may have initial warming-up or cooling, also causing the drying rate or flux to be variable in this first period. If anything remains about constant, it is the water activity, and so after 25 years I repeat my recommendation to call this first period the “Constant Activity Period” (Kerkhof, 1975).

**Sorption isotherms, some elementary considerations**

With the multitude of materials that are dried, we may expect quite some mechanisms of interaction of water and solids. We may have:
- unsaturated solutions
- saturated solutions with crystals
- supersaturated solutions
- amorphous hydrophylic solids with limited swelling capacity
- porous non-deformable solids, with various levels of porosity
- porous deformable solids, with various levels of porosity
- cellular solids

and any combination of the above. Although already a considerable number of sorption isotherms have been tabulated, there is not yet a systematic accessible database like the DECHHEMA one for fluid equilibria. There has not yet been formed a generally accepted scientific opinion which physical models and equations are the preferred descriptions for certain systems, which has been done for the other databases. Good tabulations and analysis were made on sorption isotherms of foodstuffs (e.g. Iglesias and Chirife 1982), however a digital representation and accessibility would be very welcome. In some papers...
still very exotic equations are fitted, often with high-degree polynomials involved, the tools for non-linear regression are used as a magic wand, and the sum of squares is given more attention than physics. It is horrible to see that in Arrhenius-type or power-law equations the temperature in °C is introduced instead of the absolute temperature. A lot is to be gained for drying by reading works from other areas such as adsorption. Very clear reviews about the adsorption on solids are given a.o. by Yang (1987), and Rouquerol et al. (1999).

As we have seen in Figure 2, at lower water contents we have a considerable influence of the temperature on the sorption isotherm. It is striking to see that in drying literature nearly everyone is happy with stating that we have “bound water” here, without feeling any need to specify what or where this water is. The term “bound water” has been used as a label for a multitude of different physicochemical situations, basically giving us an excuse not to define these situations, and providing the opportunity for ascribing all kinds of more or less mysterious properties or phenomena to this “bound water”. In short, it is an easy way to avoid thinking and still feel happy with each other. Let us look for a moment at the field of adsorption, or heterogeneous catalysis. During many decades a large scientific community has focussed on the presence and properties of adsorbed molecules, in relation to both structure of the adsorbent and physical properties of the adsorbates (see e.g. Kärger and Ruthven, 1992; Keil, 1999); In the first place this has led to a nice set of sorption isotherm models with associated equations. Probing the adsorbate molecules with NMR and the solid material structure with X-ray diffraction has given good insight in the spatial distribution, binding energy and mobility of the adsorbed molecules. This is matched nowadays by large efforts in molecular modeling of both equilibria and transport. The sorption isotherms of various alkanes on zeolite as evaluated from CBMC (Configurational Biased Monte Carlo) calculations are in quantitative agreement with experimental data (Vlugt et al., 1999). One of the challenges for the drying community will be to apply this methodology to a variety of drying materials. The reward is that also in the area of transport kinetics a much clearer insight on molecular scale helps us in the formulation of macroscopic models.

Some thermodynamics

Next to a host of different types of solids we also have quite some solutions or swollen matrices of carbohydrates and proteins that are dried. Here we may gain by looking at the thermodynamics of (polymer) solutions and gels, e.g. recent work by Hino and Prausnitz, 1999, Prausnitz et al., 1986. Here I want to give a simple example in order to show perspective. In a molecular mixture we have:

$$a_w = \gamma_w x_w$$  \hspace{1cm} (3)

For carbohydrates the Norrish-equation (1966) can often be used with quite some success:

$$\ln \gamma_w = k_N x_s^2$$  \hspace{1cm} (4)

What is maybe less known in the drying world is the background of such an equation. For the activity coefficient of water we have:

$$RT \ln \gamma_w = \left( \frac{\partial n_l g^E}{\partial n_w} \right)_{T, p, n_s}$$  \hspace{1cm} (5)

in which $g^E$ is the molar excess Gibbs free energy of mixing. As has been done for many vapour-liquid and liquid-liquid equilibria, one has to find expressions for $g^E$ in terms of composition, after which the activity coefficient can be derived. Next to refined models in which the expressions for $g^E$ are derived on the basis of physics, there is also a class of formal models. The simplest one is that of Porter, assuming:

$$g^E = A x_1 x_2$$  \hspace{1cm} (6)
for a binary mixture this works well e.g. for the vapour-liquid equilibrium benzene-cyclohexane. By carrying out the differentiation we obtain:

$$RT \ln \gamma_1 = Ax_2^2$$  \hspace{1cm} (7)

which is equal to the Norrish equation. In order to analyze our isotherms also for different temperatures, we have to make a little refinement. Again from basic thermodynamics we have:

$$g^E = h^E - T s^E$$  \hspace{1cm} (8)

and our easiest assumption could be that both the excess enthalpy of mixing $h^E$ and the excess entropy of mixing would obey the symmetry equation (6):

$$h^E = A_1 x_1 x_2 \hspace{1cm} s^E = A_2 x_1 x_2$$  \hspace{1cm} (9)

Then we obtain for the activity coefficient:

$$\ln \gamma_1 = \left( \frac{A_1}{RT} - \frac{A_2}{R} \right) x_2^2$$  \hspace{1cm} (10)

and for the temperature dependence of the water activity we have:

$$\left[ \frac{\partial \ln a_w}{\partial (1/T)} \right]_{p, n_w, n_s} = \left[ \frac{\partial \ln \gamma_w}{\partial (1/T)} \right]_{p, n_w, n_s} = \frac{h^E}{R} = \frac{A_1}{R} x_2^2$$  \hspace{1cm} (11)

For a long time we have worked with aqueous maltodextrin solutions, and although this is a mixture of various sugars, I felt it worthwhile to try this simple model. By fitting the data of the three temperatures an effective molecular mass for MD20 was found as 703 kg/kmol, and $A_1 = -132$ kJ/mol; $A_2 = 340$ J/mol K. In Figure 2 the drawn lines are found from this thermodynamic model, and in Figure 5 the excess mixing enthalpy and entropy are given vs. the moisture content. There are a few arguments for using this thermodynamic approach. We have also been involved in freeze-drying, and one of the physical relations of interest was the freezing curve. Assuming pure ice separates out, we have the equilibrium condition:

$$a_w = \frac{p_{ice,sat}}{p_{w,sat}}$$  \hspace{1cm} (12)

and combination with the thermodynamic data thus provides the freezing curve, as presented in Figure 6.

Another application is in multicomponent mass transfer problems. These occur in the description of aroma transport in drying extracts, and also in modeling osmotic dehydration. In general one can describe
such systems with the Maxwell-Stefan equations (Thijssen and Rulkens, 1968; Krishna and Wesseling, 1997):

\[ c_i \nabla \mu_i = -RT \sum_{j=1}^{n} \frac{N_i x_{ij} - N_j x_{ji}}{D_{ij}} \]  

(13)

Alternatively one may used the inverted equations in the multicomponent Fick form (Chandrasekaran and King, 1972):

\[ N_i = -\sum_{j=1}^{n-1} D_{ij}^F \nabla c_i \]  

(14)

Equation (13) has however the advantage that the thermodynamic factors are more clearly visible, and also that for the Maxwell-Stefan diffusion coefficients holds:

\[ D_{ij} = D_{ji} \]  

(15)

Later on we will discuss some more about the diffusion equations; let us here concentrate on the thermodynamic terms. We can write:

\[ c_i \nabla \mu_i = c_i RT \sum_{j=1}^{n-1} \left( \frac{\partial \ln a_i}{\partial x_j} \right) \nabla x_{ij} = c_i RT \sum_{j=1}^{n-1} \left( x_i \frac{\partial \ln \gamma_i}{\partial x_j} + \delta_{ij} \right) \nabla x_{ij} \]  

(16)

For ternary aroma diffusion because of the low concentrations one can treat all aroma components individually, and neglect the influence of these components on the water activity.

For osmotic dehydration by means of sugar solutions, and with a carbohydrate matrix, one could make a try with an extended symmetric model:

\[ xC_{12} = \frac{A}{x_1 x_2 + B x_1 x_3 + C x_2 x_3} \]  

(17)

which for the activity coefficient of component 1 gives:

\[ RT \ln \gamma_1 = A x_2 (1 - x_1) + B x_3 (1 - x_1) - C x_2 x_3 \]  

(18)

and similar relations can be derived for the other components. Taking water as component 1, we can see that the constants \( A \) and \( B \) follow directly from the individual sorption isotherms of the sugar and the matrix; one has to find an appropriate molecular mass however for the matrix. By soaking the matrix with different amounts of sugar, one can determine the isotherms of some ternary mixtures and in this way find the constant \( C \). Water activity measurements could be complemented by means of freezing curves, and by osmotic pressure measurements, using the relation:

\[ \Delta \Pi = \frac{RT \ln a_w}{V_w} \]  

(19)

If one would perform osmotic dehydration with salt solutions, things become considerably more complicated. One could imagine that one starts with:

\[ g^E = g^E_m + g^E_{salt} \]  

(20)

in which \( g^E_m \) is a model for the matrix-water interaction, such as treated afore, and \( g^E_{salt} \) would be modelled for the salt-water interaction, such as by Pitzer’s model (Sandler, 1989) and Bromley’s equations (1973). In their description of osmotic dehydration Yao and Le Maguer (1996,1997) used the Boyle- van ’t Hoff equation.

I have shown here some relatively simple models. For more advanced models such as UNIQUAC, which we used to describe the non-ideality of aqueous polymer solutions and others I like to point to the literature (Prausnitz et al, 1986).

In conclusion, the main intention of this paragraph was to show that doing thermodynamics like in chemical engineering might add considerably to the way we can quantify a large variety of drying processes.
Sorption isotherms, a possible pitfall

One of the items to pay attention to in determining sorption isotherms is the experimental measurement of the moisture content. Again in our own laboratory we had some complicated results when analyzing data on catalyst mixtures. One of the most common ways of doing this is by heating the material for a given time in an oven at a given temperature, or putting samples over silica in a dessicator. Assuming that time is long enough to approach equilibrium, we still have to account for the finite water activity in the oven or desiccator. If we are dealing with very hygroscopic solids such as zeolites, also at these very low water activities we still have a non-zero moisture content. In our data-unification, this might deserve attention.

Specific heat

In most drying processes the temperature is so limited that for the constituents a constant averaged specific heat may be used. Things are more complicated when phase changes occur, such as melting and solidification of fats, crystallization of solids, and recrystallization due to changes in degree of hydration. If no phase change occurs, the enthalpy on dry solids base can be written as:

\[ h' = (c_{ps} + X_w c_{pw}) \theta \]

taking \( \theta = 0 \) °C as the reference point. If we have a phase change of part of our solids, say solids nr. 2, we have:

\[ h' = (\omega_1 c_{ps1} + X_w c_{pw}) \theta + \omega_2 \left( c_{ps2} \theta + \lambda_{c,0} \right) \]

\[ \lambda_{c,0} = \lambda_{c,\theta_c} - (c_{ps2}^{high} - c_{ps2}^{low}) \theta_c \]

in which the \( \omega \) are here the mass fractions of the dry solid, and \( \lambda_{c,0} \) and \( \lambda_{c,\theta_c} \) are the latent heats of phase change at 0 and \( \theta_c \) °C respectively. The superscripts “high” and “low” indicate regions above and below the phase change temperature.

Drying Kinetics on Particle Level

Correlations

One of the methods still quite often employed is simply correlating drying curves by fitting to a suitable equation, e.g. the Page equation, or a polynomial of the water content vs. time. The coefficients can then also be fitted as a function of the air temperature, initial moisture content, particle size and whatever other process variables are of interest. This might work well for design, provided that the experimental parameters are well within the space of those to be expected for the industrial unit. It is clear that the predictive character is very low for other circumstances or materials.

One step further is the concept of the characteristic drying curve (CDC). The basic idea here is that drying curves in the falling-rate period for different temperatures and air relative humidities can be normalized to a single curve. Intersection with the curve of the constant-rate period would construct the drying curve. For the basics the reader is referred to Krischer and Kast (1978) and Keey (1972); in the latter also a good overview is given of using this concept in evaluating drying schemes. Relations with other so-called short-cut methods are treated by Coumans (2000); a limited test for the accuracy in prediction has been done by myself (Kerkhof, 1995). The satisfaction about results obtained with this method is mixed. Looking at the drying literature and the IDS proceedings, it appears that attention is shifting towards other models, more based on diffusion mechanisms. Although the concept of the CDC is relatively simple, the
work to be carried out for various dryers is still considerable and is best done by numerical solution. With
the power and price of PC’s nowadays also more intensive numerical schemes are no longer prohibitive,
and possibly this has an effect on the methods of attack.
Another method of correlation is by pseudo-scientific equations. Some equation is taken from its rightful
background, and limitations, and used for fitting the drying curve. This is OK, but often the authors claim
that this is “Fick’s Law”, or the solution from Crank or Luikov. Often they look like:

$$\frac{X - X_e}{X_{cr} - X_e} = 6 \pi^2 \exp \left( -\frac{\pi^2}{R^2} \frac{Dt}{R^2} \right)$$

(23)

and sometimes a second term is added. This type of equation has been used both for the whole of the
drying process, and for the falling-rate period. Equation (23) can be obtained only as the approximate
mathematical solution under quite a number of limiting assumptions, which most of the times are not
fulfilled in reality. As a consequence the parameter D does not really represent a material physical
property, but is found to depend on initial moisture content, air velocity, particle size and bed height. Also
one finds analyses of drying curves neglecting the equilibrium moisture content $X_e$.

\textit{External heat and mass transfer}

In considering a large number of manuscripts on drying it becomes clear that the concepts of transport
phenomena as known in chemical engineering and physics have not found recognition in all application
areas, or with all researchers. This is clearly an area in which just reading the classical textbooks may
bring improvements (Bird et al., 1960). It is possible that for a considerable part of the drying process, the
resistance to mass transfer is located mainly within the drying material. However, some transient
phenomena such as initial warming up or cooling, as well as the heat and mass transfer rate in the
constant activity period are determined by the heat and mass film transfer coefficients. Moreover the
internal concentration profiles as they build up during this initial period are dependent on the external
heat and mass transfer coefficients.

For the convective drying of sheets or stacks of larger units like bricks, the transfer coefficients vary with
the location according to boundary layer theory. For many practical drying processes the correlations
from chemical engineering provide sufficient accuracy; for more complicated geometries modelling with
CFD will give accurate results about local coefficients (see also Menshutina and Kudra, 2000). It is of
interest to publish such data in the form of local $Sh$ and $Nu$ numbers in dependence on $Re$ next to the
whole of the specific solution (Esfaharian et al., 1998). This constitutes a nice addition to the classic work
in this area (Krischer and Kast, 1978).

In any case the effect of non-equimolar transport should be taken into account, as given by Bird et al.
(1960; see also Kerkhof and Schoeber, 1974).

Also for fluidized and spouted beds sufficiently accurate transfer equations are available. One should take
care however to distinguish between local coefficients on the particle level, and bed-averaged coefficients
(see e.g. Schlünder and Tsotsas,1988)

\textit{Interlude: Idealized Diffusion in a Sphere}

As so very often more or less simplified equations are quoted and (mis)used I make a small stop here
to review the problem of diffusion in a sphere. The problem and solutions and many more things have
been presented in the classic textbooks (Carslaw and Jaeger,1959, Crank, 1975, Luikov, 1968). For many
readers this will also be standard material; however in an effort of unification it might help to highlight
some points.

Let us assume that we have isotropic diffusion in a sphere. The differential equation for this reads:

$$\frac{\partial c}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( Dr^2 \frac{\partial c}{\partial r} \right)$$

(24)
For the case of a constant diffusion coefficient this can be written as:

\[
\frac{\partial c}{\partial t} = \frac{D}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial c}{\partial r} \right)
\]  

(25)

In order to solve this equation one needs one initial and two boundary conditions. For the initial condition we can write in general:

\[
t = 0 \quad 0 < r < R \quad c = f_0(r)
\]

(26)
in which \(f_0(r)\) is the initial distribution. In the center of the sphere we assume symmetry:

\[
t > 0 \quad r = 0 \quad \frac{\partial c}{\partial r} = 0
\]

(27)

Related to drying might be two cases might be of main interest, the constant-rate period, and the falling rate period after that. For the constant-rate period we may assume a constant drying flux, and a uniform initial distribution:

\[
t = 0 \quad 0 < r < R \quad f_0(r) = c_0
\]

\[
t > 0 \quad r = R \quad -D \frac{\partial c}{\partial r} = N = \text{constant}
\]

(28)

Note that we have also assumed that the sphere radius does not change. The mathematical solution is (Crank, 1975):

\[
c_0 - c = \frac{NR}{D} \left[ \frac{3Drt + 1}{2R^2} - \frac{3}{10} - 2R \sum_{n=1}^{\infty} \frac{\sin(\beta_n r)}{\beta_n^2 R^2 \sin(\beta_n R)} \exp(-D\beta_n^2 r) \right]
\]

(29)
in which the \(\beta_n\) are the roots of the characteristic equation:

\[
\beta_n R \cot(\beta_n R) = 1
\]

(30)

We can find the roots in Crank, \(\beta_1 R = 4.4934, \beta_2 R = 7.7253, \ldots\) and so we see that we may have a long time solution in which the series term disappears, and we would have a development towards a parabolic profile, finally uniformly moving with time. If we keep on evaporating water at a constant rate it is clear that the solution at some point will predict negative concentrations. As follows from the sorption isotherm we may want to define a critical surface water concentration at which the constant activity period stops, say \(c_{cr}\) and so the length of this period, under all assumptions made, would be given by:

\[
c_0 - c_{cr} = \frac{NR}{D} \left[ \frac{3D_{ca}t + 1}{2R^2} - \frac{3}{10} - 2R \sum_{n=1}^{\infty} \frac{1}{\beta_n^2 R^2} \exp(-D\beta_n^2 t) \right]
\]

(31)

So we may end with a parabolic concentration profile, if the flux is low enough, or rather if the flux parameter \(NR/(Dc_0)\) (related to the Kirpichev number) is low enough. If we have a high value of this flux parameter, the profile will not have penetrated to the heart of the sphere when the critical surface concentration is reached. For short times an approximate solution is (Luikov, 1968):

\[
c_0 - c = \frac{NR}{D} \left[ \exp \left( \frac{Fo - R - r}{R} \right) \text{erf} \left( \frac{1 - r / R}{2Fo^{1/2}} - Fo^{1/2} \right) - \text{erf} \left( \frac{1 - r / R}{2Fo^{1/2}} \right) \right]
\]

(32)

So, also from this we can estimate the length of the CA-period.

Now we are ready to proceed with the falling rate period. This means that we have to change boundary condition; however in order to obtain an analytical solution we have to solve the differential equation again, with a new starting profile:

\[
t' = t - t_{ca} = 0 \quad 0 < r < R \quad c = f_{ca}(r)
\]

(33)
in which we have to choose which one of (29) or (32) we think is more appropriate. Then we have to define the new boundary condition. In principle we have here:
\[ t > 0 \quad r = R \quad - D \frac{\partial c}{\partial r} = k_f \left( c^i - c^b_c \right) \] (34)
in which \( c^i_c \) is the continuous phase (air) concentration at the interface, in equilibrium with the concentration at \( r = R \) inside the sphere. The only way to obtain an analytical solution with this boundary condition is to assume a linear equilibrium relationship:

\[ c^+_c = mc \] (35)

In that case the solution is given by:

\[ c - c^{b*} = \sum_{n=1}^{\infty} \frac{2\mu_n}{\mu_n - \sin \mu_n \cos \mu_n} \sin(\mu_n r / R) \int_{r=0}^{r=R} f_{ca}(r) \sin(\mu_n r / R) dr \exp\left( -\mu_n^2 Fo \right) \] (36)
in which the eigenvalues \( \mu_n \) follow from the characteristic equation:

\[ \tan \mu = \frac{1}{1 - Bi_m} \mu \] (37)

Here \( c^{b*} \) is the concentration in the sphere in equilibrium with the continuous phase bulk concentration. So the solution depends on the Biot number for mass transfer:

\[ Bi_m = \frac{k_f R m}{D} \] (38)

Note that for such problems it is essential that in the Biot number the slope of the equilibrium line is included.

Now, what does this all contribute to unification of our theories? In the first place, the foregoing shows how the mathematics are, under a number of strict assumptions: \( D \) and \( R \) constant, \( N \) constant, and in the falling rate period \( m \) and \( c^b_c \) also constant. (It is also possible to derive an analytical solution for time-variable \( c^b_c \), however it looks already complicated enough). Further we see that the concentration profile after the constant activity period is the starting point for the next period.

Now we may try some further simplification. One could be that we assume that the CA-period is so small that we start immediately in the falling-rate period, and we take the initial profile as equal to \( f_0(r) = C_0 \).

Then the solution would be:

\[ \frac{c_0 - c}{c_0 - c^{b*}} = 1 - \sum_{n=1}^{\infty} (-1)^{n+1} \frac{2Bi_m \left( R^2 + (Bi_m - 1)^2 \right)^{1/2}}{\mu_n^2 + Bi_m (Bi_m - 1)} R \sin(\mu_n r / R) \exp\left( -\mu_n^2 Fo \right) \] (39)

If we would further assume that all mass transfer resistance is inside the sphere, \( Bi_m \rightarrow \infty \), we have \( \mu_n = n\pi \), and the solution reads:

\[ \frac{c_0 - c}{c_0 - c^{b*}} = 1 - \sum_{n=1}^{\infty} (-1)^{n+1} \frac{2 \sin(\mu_n r / R) \exp\left( -\mu_n^2 Fo \right)}{\mu_n^2} \] (40)

and, only for this situation, integration over the particle volume gives for the average concentration:

\[ \frac{c_0 - c}{c_0 - c^{b*}} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left( -\mu_n^2 Fo \right) \] (41)

which for sufficiently long time, \( Fo > 0.05 \), can be well approached by a single term of the series.

So this was for an immediate falling-rate period, again with \( D, R \) and \( c^b_c \), with \( Bi_m \rightarrow \infty \). Another approximation that we can make, is to take into account the CA-period, but neglect the profile at the end of this period, and approximate by
It depends on the steepness of the profile after the CA-period how large the error is. If we compare the assumptions made with drying practice we may note the following:
- In drying the isotherm can only approximately be split up in a horizontal and a sloped part.
- During drying the particle temperature changes often considerably. This means that the diffusion coefficient will change, as well as the slope of the equilibrium curve:

$$m = \frac{p_{w,\text{sat}}}{RT} \frac{da_w}{dc_w}$$

(43)

- For most materials the diffusion coefficient depends on the moisture content.
- For particles moving through a dryer, the humidity and temperature of the drying air they see in the bulk, is variable.

In conclusion, the application of series solutions is very limited, and if done, the assumptions underlying the analytical solution should be verified in detail.

Internal mass and heat diffusion: Descriptive equations

As stated in the consideration of sorption isotherms, there are many ways in which water can be present, consequently there are also many mechanisms by which water transport can occur. Relatively simple systems are food solutions and non-porous films; here we have in general molecular diffusion as the mass transfer mechanism, and generally the temperature gradients in such particles are negligible. For many applications one may work with lumped-mechanism approximations in which use is made of an effective diffusion coefficient for moisture. In the most general form we have:

$$\mathbf{j}_w = -L_{mm} \nabla \rho_w - L_{mT} \nabla T - L_{mp} \nabla p$$
$$\mathbf{j}_q = -L_{qm} \nabla \rho_w - L_{qT} \nabla T - L_{qp} \nabla p$$

(44)

for the heat and mass fluxes, provided that the pressure gradient is independent of the concentration and temperature gradients. In drying of porous materials generally the total pressure gradient is dependent on the other gradients, and the flux equations reduce to:

$$\mathbf{j}_w = -L_{mm} \nabla \rho_w - L_{mT} \nabla T$$
$$\mathbf{j}_q = -L_{qm} \nabla \rho_w - L_{qT} \nabla T$$

(45)

as presented long ago by e.g. Luikov and Mikhailov (1961), Philip and de Vries (1957). The coefficients $L_{\alpha\beta}$ can be called phenomenological; a lot of scientific progress has been made to build them up from mechanistic descriptions and suitable volume averaging methods (Whitaker, 1998; Couture et al, 1995; Coumans and Ramakers, 2000), and within the contributions to this symposium we see also experimental work on determining the moisture and concentration profiles.

It appears that for most problems the equations of change can be given by the continuity equations:

$$\frac{\partial \rho_w}{\partial t} = - (\nabla \cdot \mathbf{j}_w)$$
$$\frac{\partial h'}{\partial t} = - (\nabla \cdot \mathbf{j}_q)$$

(46)

thus leading to parabolic differential equations. However, as also in chemical engineering, there is a minority proposing hyperbolic equations of the type:

$$\frac{\partial Z}{\partial t} + \tau_Z \frac{\partial^2 Z}{\partial t^2} = - (\nabla \cdot \mathbf{j}_Z)$$

(47)

with $Z$ for concentration or temperature (Kronberg et al.,1998; Nastaj, 2000). It would be valuable to perform a critical theoretical and experimental investigation what framework to use and when.
It is clear that in situations with large temperature gradients, such as in contact paper drying, one has to use equations (45), and preferably with mechanistic models with independently estimated parameters. For moderate temperatures in convective drying often the internal temperature gradients can be neglected, and we obtain the isothermal diffusion equation:

\[ j_w = -D_{eff} \nabla \rho_w \]  

(48)

and for many products the effective diffusion coefficient can be regarded as a material property, depending however on moisture content and temperature. The relevance of taking into account temperature gradients can be estimated from the thermal Biot-number:

\[ Bi_q = \frac{\alpha R}{\lambda} \]  

(49)

and for particle drying this is often in the order of \( Bi_q \approx 0.1 \), indicating that most of the resistance for heat transfer is located in the air phase.

Equation (48) has been applied for the description of drying hollow and non-hollow droplets (van der Lijn, 1976; Kerkhof and Schoeber, 1974), including the effect of shrinkage. Also it can be well applied for the drying of biomaterials (Lievense et al., 1990; Kerkhof, 2000), as shown in Figures 7 and 8.

The experimental determination of the effective water diffusion coefficient as a function of concentration from drying curves follows from the Regular Regime approach of Schoeber, Liou, Coumans, as reviewed by Kerkhof (1994) and Coumans (2000).

In the above the solids is treated as one single pseudocomponent. Meerdink et al. (1995) studied the segregation of protein and sugar during drying, and developed a ternary diffusion model for this. For the osmotic drying with two solids one could write equation (13) and (16) combined as:

\[ \Gamma_{11} \nabla x_1 + \Gamma_{12} \nabla x_2 = \frac{N_{2}x_1 - N_{1}x_2}{c_1D_{12}} + \frac{N_{3}x_1 - N_{1}x_3}{c_1D_{13}} \]

\[ \Gamma_{21} \nabla x_1 + \Gamma_{22} \nabla x_2 = \frac{N_{1}x_2 - N_{2}x_1}{c_1D_{12}} + \frac{N_{3}x_2 - N_{2}x_3}{c_1D_{23}} \]  

(50)
with
\[ \Gamma_{ij} = x_i \frac{\partial \ln \gamma_i}{\partial x_j} + \delta_{ij} \] (51)
is the so-called thermodynamic factor (Krishna and Wesselingh, 1997). Taking water as component 1, and sugar as component 2, we have the matrix solids as 3. As discussed before we can estimate the thermodynamic factors, and in first approximation we would take for \( D_{12} \) and \( D_{13} \) the binary diffusion coefficients as they can be found for water-sugar and water-matrix. We should note however that in our experiments we determine the Fickian diffusion coefficient, and so we have to evaluate the Maxwell-Stefan diffusion coefficients from the binary system equation:
\[ D_{12}^F = D_{12} \left( 1 + \frac{\partial \ln \gamma_1}{\partial x_1} \right) \] (52)
and a similar relation for \( D_{13} \). It is to be expected that all diffusion coefficients are dependent on concentration. We can invert the equations (50) to give formal expressions for the fluxes, and solve the continuity equations with the appropriate boundary conditions. By comparison with experimentally determined concentration profiles and fluxes then a relation between \( D_{23} \) and composition can be estimated. It should be noted that the set of equations, given here on mole fraction can be converted also to molar and weight concentration basis. The formulation of the boundary conditions requires knowledge about the physics of the system: does the material shrink, do we have a convective condition in the continuous phase, is the continuous phase a well-mixed bath or are we using countercurrent flow?
It is not my intention to solve all these problems here, but rather put attention to the generality of this type of modelling.

Some aspects of transport in porous media

Many drying materials are heterogeneous porous systems, some also non-isotropic and sometimes deformable. For a good description of such systems we need to know the governing transport mechanisms in the various subregions of interest, to formulate the correct and adequate averaging rules of both transport rates and driving forces over different representative volume parts of our medium (Whitaker, 1977, 1998). For deforming systems also mechanical deformation models need be established, including constitutive relationships, and the influence of compressive and expansive stresses on mass transport and energy dissipation. Next to this also the parameters of the various models should be determined from experiments. Excellent work is presented by Perré and May, 2000; Perré and Turner, 2000; Kowalski, 2000. Prat (2000) provides an extensive overview of the way water is present in porous networks, and the effect on drying. For osmotic drying models were set up taking onto account cell structure and membranes (Yao and Le Maguer, 1996, 1997).

The modeling of transport in pores, pore networks and fractal geometries is a field of common interest with heterogeneous catalysis and adsorption. Recent overviews for catalysis are given by Keil (1999) and Rieckmann and Keil (1999), of which the methodology may have considerable merit also for drying. Both in detailed and in averaging models one has to use descriptions for the motion of water in the different phases. I would like to focus here on two areas, in which again the integration with knowledge from other areas may be fruitful, the transport of vapour and air, and the transport of bound water. In most
models, also presented here at IDS, the transport of vapour is treated as a convection-diffusion problem of the kind:

\[
N_{wv} = N_{gt}x_{wv} - D_{wg}c_t \frac{\partial x_{wv}}{\partial z} \\
N_{gt} = -c_t \frac{K_g \partial p_{gt}}{\eta_g \partial z} 
\]

with adequate corrections for tortuosity and porosity. In case the air is assumed to be stagnant this leads to the Stefan-equation. In the field of liquid membrane ultrafiltration and gas transport in mesoporous media considerable attention has been given to the effects of wall friction of the individual components. In drying this seems to draw only limited attention (Hallström, 1985). In recent work we developed novel models to describe such phenomena. For the pore-averaged fluxes in a single capillary we approached with the binary friction model (Kerkhof, 1996):

\[
\frac{\partial p_w}{\partial z} = RT \frac{x_wN_a - x_aN_w}{p_tD_{wa}} - f_{wm}RTN_w \\
\frac{\partial p_a}{\partial z} = -RT \frac{x_wN_a - x_aN_w}{p_tD_{wa}} - f_{am}RTN_a 
\]

in which the wall friction coefficients are given by:

\[
f_{im} = \left(D^K_i + \frac{n_i^0}{8\kappa_i} \right)^{-1} 
\]

in which \(D^K_i\) is the Knudsen diffusion coefficient of \(i\), and \(\kappa_i\) is the fractional viscosity coefficient of \(i\):

\[
\kappa_i = \frac{1}{p_t} \sum_{j=1}^{n} x_j \xi_{ij} \\
\xi_{ij} = \left[1 + \left(\frac{\eta_j^0}{\eta_j^0} \right)^{1/2} \left(M_j / M_i \right)^{1/4} \right]^2 \\
\xi_{ij} = \frac{\left[1 + \left(\frac{\eta_j^0}{\eta_j^0} \right)^{1/2} \left(M_j / M_i \right)^{1/4} \right]^2}{8\left[1 + M_j / M_i \right]} 
\]

From calculations for air-water follows that at atmospheric total pressure these effects become considerable for pores of 1 µm and smaller, and so the convective-diffusion equation (53) does not hold any more (Kerkhof, 1997). Based on a precise component momentum balance we derived very recently the velocity profile model for the transport in pores (Kerkhof et al., 2000). In Figure 10 the velocity profiles are given for the case of the Stefan-tube, so assuming stagnant air. For very small pores we have Knudsen flow and the velocity profiles are flat. For larger pores we see clearly that in the center the water vapour has a high velocity, and it drags along the air.
molecules there. Since there is no net average air flow assumed here, there has to be a backflow of air near the walls of the pore. Also we see that there is slip at the wall.

In modeling the transport of bound water, also in most papers it is not specified what or how the water is bound, and a simple Fickian type diffusion is assumed. In minerals the bound water can be assumed to be present in micropores, and so we may apply micropore or surface diffusion. In earlier work this has been studied e.g. by Toei et al. (1983). In the description of multicomponent adsorption a Maxwell-Stefan theory was recently developed by Krishna (1990), which for water transport can be written as:

\[
N_{wb}^s = -c_{sat}^s D_s \Gamma \frac{d\theta_w}{dz}
\]

\[
\Gamma = \theta_w \frac{\partial \ln a_w}{\partial \theta_w}
\]

(57)

Recently Silva et al. introduced both the binary friction model and Krishna’s micropore diffusion theory in the estimation method for effective diffusivity in drying (Silva et al, 2000).

So again we see that we can use theoretical concepts from different fields in drying.

Numerical solutions

With the present state of the art, the numerical solution of the transient diffusion equations can be performed quite well on a PC, at high speed, and even multi-particle simulations are feasible. On the particle level very often we use finite-difference methods when the problem has been approached as one-dimensional, of a modified Crank-Nicholson type. For more-dimensional problems finite-element and finite-volume methods have been used, analogous to methods in CFD, see (Perré and May, 2000. Less known are methods that are used in chemical engineering. One is the writing the problems in terms of simultaneous first-order differential equations, and solving them by appropriate integration routines; available are the packages based on the method of lines (Schiesser, 1991), which is also implemented in the NAGLIB (1998). Collocation methods in chemical engineering are well-developed, see Villadsen and Michelsen (1978). Here important computation time savings can be obtained, however still research has to be done about accuracy and optimal weighing functions. Regular Regime methods have been reviewed recently by Coumans (2000); for low driving force situations still development is necessary (Kerkhof,1995).

EQUIPMENT AND PLANT LEVEL

In the modeling of kinetics at the equipment level one needs to set up the equations of change for the gas phase and the particles or product units. Depending on the operation hydrodynamics may play an important role, such as in spray drying, and CFD modeling is increasingly used also in drying simulations. The capabilities of the packages have improved, but especially the capacity of workstations and supercomputers allow us now to perform very calculation-intensive problems. One of the challenges is to implement rigorous drying kinetics into such distributed models. In our work on modeling of spray drying we implemented regular regime short-cut calculations for the drying rate in all our particles, and so we could combine this with the CFD two-way coupled model. In our analysis of thermal degradation we took all recorded histories of the particles surroundings and solved the diffusion equations together with the degradation reaction, and by integration over the particle distribution we obtained the mass-averaged quality (Kieviet and Kerkhof, 1996). As discussed above, it is desirable to implement good particle kinetics. Fyhr et al. (1999) have set up multi-particle models for continuous fluid-bed drying, with a stochastic model for the particle trajectory. In Figure 11 results are shown of a simulation of a fixed bed drier, divided in 25 layers, in which for each layer the diffusion equation was solved, together with the air mass and enthalpy balances. In such calculations it would still be desirable to have fast short-cut methods.
In chemical engineering Hoomans et al. (1996) have modeled and measured multi-particle motion; combination of such work with drying kinetics is a challenging perspective. It would be interesting to compare results and methodology such as in Figure 11 with the theory of composition and thermal waves in fixed-bed adsorbers (Yang, 1987), and find out whether phenomena such as constant patterns can also be found in drying. Finally the development of flow-sheeting programs for dryers with all kinds of products can be carried out with the methodology of that in the chemical industry. Process control and optimisation requires the introduction of more gray and white box models next to the implementation of modern methods such as fuzzy logics and neural networks.

CONCLUDING REMARKS

I hope to have demonstrated that drying science and resulting design and operation rules can gain from cross-breeding with various branches of science, and that a common framework can be set in this way. On one hand it may require some reorientation, on the other hand it may be also be rewarding, as shown in the perspectives of applying more thermodynamics. Big challenges for the drying community will be to set up a joint knowledge framework and a methodology to process information. It would be very desirable to have access to a drying database, in which carefully reviewed theories and data could be accessed, preventing as much as possible the reinvention of the wheel. This requires a cooperative effort of scientists, end-users and consultants, and additional funding which we will have to find.

ACKNOWLEDGEMENT

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NOTATION

Roman symbols:

\( A \) surface area \( m^2 \)
\( A,B,C \) mixture parameter (Equations (6),(17)) \( J/kmol \)
\( a \) thermodynamic activity -
\( Bi \) Biot number -
\( c \) molar concentration \( kmol/m^3 \)
\( c_p \) heat capacity \( kJ/kg K \)
\( D \) Fickian diffusion coefficient \( m^2/s \)
\( D \) Maxwell-Stefan diffusion coefficient \( m^2/s \)
\( f \) concentration distribution \( kmol/m^3 \)
\( f_{im} \) friction coefficient of \( i \) with wall \( \text{s/m}^2 \)

\( Fo \) Fourier number \( Dt/R^2 \) -

\( g \) Gibbs free energy \( \text{J/kmol} \)

\( H \) absolute humidity kg water/kg dry matter

\( h, h' \) enthalpy \( \text{J/kmol, resp. J/kg} \)

\( j, j' \) mass, heat flux \( \text{kg/m}^2 \text{s, W/m}^2 \)

\( j \) counting variable -

\( K \) permeability \( \text{m}^2 \)

\( k_N \) Norrish constant (Equation (4)) -

\( L_{oij} \) phenomenological coefficient -

\( M \) molecular mass \( \text{kg/kmol} \)

\( m \) slope of equilibrium line -

\( N \) molar flux \( \text{kmol/m}^2 \text{s} \)

\( n \) number of moles, components -

\( p \) pressure \( \text{Pa} \)

\( R \) gas constant \( \text{J/kmol K} \)

\( R \) radius \( \text{m} \)

\( r \) radial coordinate \( \text{m} \)

\( r_0 \) pore radius \( \text{m} \)

\( RH \) relative humidity \( \% \)

\( s \) entropy \( \text{J/kmol K} \)

\( T \) temperature \( \text{K} \)

\( t, t' \) time \( \text{s} \)

\( \bar{V} \) specific volume \( \text{m}^3/\text{kmol} \)

\( W \) mass \( \text{kg} \)

\( X \) water content on dry basis kg water/kg dry solids

\( x \) mole fraction -

\( Z \) formal variable (Equation (47)) -

\( z \) distance coordinate \( \text{m} \)

**Greek Symbols**

\( \alpha \) external heat transfer coefficient \( \text{W/m}^2 \)

\( \beta_n \) eigenvalue -

\( \Delta \) difference -

\( \delta_{ij} \) Kronecker delta -

\( \epsilon \) porosity \( \text{m}^3/\text{m}^3 \)

\( \Gamma \) thermodynamic factor -

\( \gamma \) activity coefficient -

\( \eta \) dynamic viscosity \( \text{Pa s} \)

\( \kappa \) fractional viscosity coefficient \( \text{s} \)

\( \lambda \) thermal conductivity \( \text{W/m}^2 \text{K} \)

\( \lambda \) heat of phase change \( \text{J/kmol} \)

\( \mu \) chemical potential \( \text{J/kmol} \)

\( \mu_n \) eigenvalue -

\( \Pi \) osmotic pressure \( \text{Pa} \)

\( \theta \) temperature \( ^\circ \text{C} \)

\( \theta \) occupancy of surface -

\( \rho \) concentration on weight basis \( \text{kg/m}^3 \)

\( \tau \) relaxation time \( \text{s} \)

\( \omega \) mass fraction kg/kg total

\( \xi \) Wilke interaction parameter -

\( < > \) averaged -
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
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<tr>
<td>∂</td>
<td>partial derivative</td>
<td></td>
</tr>
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<td>∇</td>
<td>gradient, divergence</td>
<td>m⁻¹</td>
</tr>
<tr>
<td>.</td>
<td>dot product</td>
<td></td>
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**Superscripts**
- 0: pure component
- b: bulk
- E: excess
- F: Fickian
- i: interface
- s: solids, surface
- *: equilibrium

**Subscripts**
- 0: initial condition, at 0 °C
- 1,2,3: component 1,2,3
- 1,2: term number
- c: phase change
- ce: continuous phase
- ca: constant activity
- cr: critical
- e: equilibrium
- eff: effective
- f: film
- g: gas phase
- i,j: component i, j
- ice: ice
- m: matrix, mass
- n: number of moles
- p: particle, pressure
- q: thermal
- s: solid
- salt: salt
- sat: saturation
- T: temperature
- t: total
- vz: vapour
- w: water

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