Preface

The work described in this report is the result of my own research, unaided except as specifically acknowledged in the text, and it does not contain material that has already been used to any substantial extent for a comparable purpose. This report contains 40 pages.

Student

Date

I confirm that the student above has cleared the laboratory space used in this project to my satisfaction.

Supervisor

Date

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Summary

Spray drying is a unit operation ubiquitous in a wide range of industries, uniquely combining product drying with particle formation. The physical processes occurring within such units are complex and this largely explains the modest fidelity of spray dryer models as compared with those of other drying technologies. The aim of this work is to produce a new spray dryer model, validated against industrial data.

This report first gives an overview of the operation of a generic spray drying unit. A review of existing dryer models is then presented, structured around the idea of a hierarchy of modelling levels. It is observed that the purpose of the model and problem to be solved will determine the appropriate choice of modelling level. For the most detailed simulations, the key processes to be modelled are identified as: the fluid flow field; particle drying; particle-particle interactions; and particle interactions with the wall. Of these, it is concluded that particle drying is fundamental and hence a detailed review of particle drying models is conducted.

Work towards a new model is introduced, where a system comprising a solution with suspended solids is considered. Evaporation, diffusion, solid formation and bubble formation will all be considered. So far, work has focussed on developing a modelling architecture and formulating the equations to describe the diffusion processes. The report concludes with a consideration of further work.
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Report produced in $\LaTeX$
1 Introduction

The spray drying process has a broad spectrum of industrial applications, from pharmaceutical to milk powder, detergents to coffee. It is unique among drying technologies in that it simultaneously accomplishes the tasks of drying and powder formation. It has the ability to accept a liquid feed and produce dry powders with specific size and moisture distributions, (Masters; 1992). Such is the importance of these factors that for many applications, despite a lower thermal efficiency than competing dryers, spray drying is the only realistic choice.

Given the ubiquitous nature of the technology, it is perhaps surprising that theoretical models of such driers are relatively poorly advanced. Historically this can be attributed to difficulties modelling the complex flow patterns and multi-phase interactions occurring within such units. However, advances in computing power, and in particular the advent of computational fluid dynamics (CFD), means that detailed simulation is now a possibility. In partnership with industry, the ultimate aim of this project is to develop a new spray dryer model, verified against data from industrial units. Such a model would be of considerable diagnostic use for analyzing existing dryer operation and also in the design and optimisation of new facilities.

This report gives background, some details of the work completed so far and an idea of the way ahead. A comprehensive survey of the spray drying literature is conducted and the main features required in a reasonably sophisticated spray dryer model identified. Justification is made for considering the particle drying model as most fundamental and for that reason the majority of the report focuses on single particle drying. A review of approaches adopted to modelling single particle drying leads the author to decide on a modelling strategy and some preliminary work in this direction is presented.

2 Spray Drying

2.1 Process Overview

Before a system can be modelled, it is clearly essential to have a good understanding of how the process works. The range of spray drying technology in current usage reflects the diversity of the industries that dry their products in this way. What follows is, therefore, only a brief overview of a generic spray drying process.

At its simplest level, spray drying involves the feed, in liquid or slurry form, being sprayed into a drying medium. This is normally hot air. In more detail, the spray drying unit operation may be considered as being composed of four stages, Figure 1.

The atomisation of a pumpable feed to form a spray is the key characteristic of spray drying. Two principle types of atomiser are used in industry: rotary atomisers making use of centrifugal energy; and pressure nozzles which exploit pressure energy to atomise the spray. Multiple injection levels may be used to handle higher flowrates. Whichever type is chosen, the initial droplet diameter will be in the range 20-500 $\mu m$. The result of the atomisation must be a spray which provides optimum evaporation conditions leading to the desired characteristics in the dried product.

The manner in which the spray droplets contact the drying medium determines their subsequent drying behaviour and, in turn, greatly influences the properties of the final product. The form of spray-air contact is determined by the location of the atomiser relative to the air inlet. Broadly speaking, the flow may be considered either ‘co-’ or ‘counter-current’.

In the co-current arrangement, the product and air pass through the dryer in the same direction. This is by far the most common arrangement, (Zbicinski and Zietara; 2004), and is
especially suited to the drying of heat sensitive products. As the wet feed immediately contacts
the hottest air, drying is rapid and the drying air cools accordingly. The product temperature
remains around the wet bulb temperature throughout the initial drying period. Subsequently,
the product is in contact with cooler air and is at no point subject to thermal degradation.
Counter-current operation offers greater thermal efficiency as the liquid feed and air enter at
opposite ends of the drier. However, this means that the driest material is exposed to the hottest
air. Consequently, the set-up is only suitable for products which are non-heat-sensitive. There
are also dryer designs which combine co- and counter-current flow patterns, termed mixed-flow
driers.

The choice of how to contact the spray with the drying air will be determined by the material
being dried and the desired product properties. Co- and counter-current arrangements give
different particle morphologies due to the different particle temperature histories. This can
lead to counter-current set ups producing a less porous product with higher bulk density.
Slower evaporation reduces the tendency to puff, lowering the particle porosity. However, as
mentioned above, the configuration may only be used for products which can withstand heat
treatment. Conversely, co-current driers feature rapid evaporation preventing high particle
temperatures. However, such high drying rates will more often lead to particle expansion
or fracture, producing non-spherical, porous particles. The degree of agglomeration will also
be effected by the dryer arrangement and, in turn, will influence product properties such as
coarseness. The morphological development of particles is considered further in Section 3.3.

Evaporation occurs as the fluid feed comes into contact with the drying air. As discussed
previously, the drying rate will be determined, in part, by the physical layout of the spray
drying operation. However, it is also important to understand how the composition of the
feed will affect the drying behaviour of the particles in the spray. The unique feature of spray
drying is that drying and formation of a powdered product occur simultaneously. Hence an
understanding of the physics of particle drying is essential for describing the unit operation.

Once the dried product has been formed, a final separation stage is necessary. Two principle
systems may be identified. In the first, (Figure 2a), primary separation occurs in the drying
tower itself, with the majority of the product being removed from the base of the tower. The
remaining product exits entrained in a separate air discharge stream, which is sent to secondary
separation equipment, e.g., cyclones, bag filters or electrostatic precipitators, (Perry and Green;
1997). The second system, (Figure 2b) operates with total recovery of the dried product in

Figure 1: The principle process stages of a generic spray drying process, adapted from Masters (1992)
the separation equipment. This places great importance on the efficiency of the separation system employed and, for obvious reasons, can only be used with a co-current set-up.

![Diagram of spray drying process](image)

**Figure 2:** Product discharge from a co-current drying system with: (a) primary separation in the drying tower; and (b) total recovery in the dedicated separation equipment. Diagram adapted from Masters (1992).

### 2.2 Dryer Simulation

The scope of the current project is limited to the simulation of the spray drying tower. Consequently, no attempt will be made to model the separation systems and these are not considered further. However, any model must address the three remaining process stages illustrated in Figure 1. What follows is an overview of the current state of spray dryer modelling.

#### 2.2.1 A Hierarchy of Modelling Levels

Despite the importance and wide application of spray drying technology, the theoretical modelling of spray dryers is still relatively poorly developed. Spray dryers are more difficult to model than other dryer types and simple scale up techniques can not be used. Instead, Oakley (2004) identifies a hierarchy of ‘modelling levels’ for simulating spray drying systems:

- **Level 0** Heat and Mass Balances
- **Level 1** Equilibrium Based Models
- **Level 2A** Rate-based with simplifying assumptions about fluid flow
- **Level 2B** Rate-based with simulation (CFD) of the continuous gas phase and particle motion.

The most appropriate choice of modelling level to use depends on the detail and accuracy required from the solution, *i.e.*, on the purpose of the model and problem to be solved.
2.2.2 Heat and Mass Balances

The simplest modelling layer is to apply appropriate energy and mass balance equations to the system. Such models require no detailed knowledge of the dryer geometry, or the processes occurring therein. However, the inlet streams must be specified, along with the exit moisture content. This limits the predictive power of such models. Nevertheless, they are useful at the earliest design stage to assess the thermodynamic feasibility of the proposed operation.

Example

As an illustration, consider the following example: it is sought to use a co-current device to dry 1800 kg/hr of product containing 32.8% moisture on a wet basis. The required outlet moisture content is 7%, again on a wet basis. It is checked whether co-current contacting with a hot air stream at 300°C, absolute humidity of 0.05 kg/kg and flow rate of 4500 kg/hr will be sufficient to achieve this duty. Losses of 50 kW are assumed, although it would be equally possible to define a unit efficiency. All the information required for the heat and mass balances is summarised in Table 1.

It is found that the proposed system satisfies the heat and mass balance equations, giving an outlet gas temperature of 74°C and relative humidity of 30%. It has been assumed in the calculation that the outlet product is at the temperature of the surrounding gas.

As observed above, the heat and mass balance equations require that the outlet product moisture content is specified to close the equation system. An alternative is to make use of phase equilibrium relationships to relate the moisture content of the product at the outlet to the humidity of the surrounding gas. Before proceeding, it will prove helpful to review the thermodynamics of drying and introduce some of the notation and terms used later in the report.

2.2.3 The Thermodynamics of Drying

Drying is the removal of moisture from a product. Materials containing moisture may be divided into two categories: hygroscopic and non-hygrosopic. Examples of hygroscopic materials are...
salts, vegetal fibres, most metal oxides and many polymers. Metal powders and glass granules are examples of non-hygroscopic products. In hygroscopic materials, moisture may be present in bound and unbound forms. Bound moisture has a water activity less than unity. The activity of water is defined

$$a_w = \frac{p}{p_{sat}},$$

where $p$ is the partial pressure of water calculated at the wet bulb temperature. $p_{sat}$ is the saturation pressure, the partial pressure of water vapour above pure water at the product temperature. Bound moisture therefore exerts an equilibrium vapour pressure lower than the partial pressure above pure water at the product temperature. Hygroscopic products may absorb water from the environment in a number of different ways, e.g., sorption with formation of a hydrate, binding by surface energy, diffusion of water into the material structure, capillary condensation, or formation of a solution. Water gained in all of these ways may represent bound moisture, with the strength with which the absorbed water is bound to the product depending upon the method of absorption.

Moisture in excess of the bound moisture is termed unbound. In a non-hygroscopic material, all moisture is unbound. Unbound moisture exerts an equilibrium partial pressure equal to the partial pressure above pure water at the same temperature, i.e., the activity of unbound moisture is unity. These terms are illustrated in Figure 3. Note that these descriptions assume pure moisture in a solid material; for mixtures, the vapour pressure will be lowered as given by, for example, the Antoine equation, (Sandler; 1998).

Humidity is a measure of the moisture content of a gas and is defined as the ratio of the mass of water vapour to the mass of vapour-free, ‘bone-dry’ gas,

$$H = \frac{\text{Mass of Vapour}}{\text{Mass of Vapour} - \text{Free Gas}}.$$
This can be written as

$$\mathcal{H} = \frac{pW_A}{(P - p)W_{\text{gas}}}$$,

where \(W_A\) is the molar mass of the solvent and \(W_{\text{gas}}\) is the molar mass of the gas. \(P\) is the total pressure and \(p\) is the moisture partial pressure. The equilibrium relative humidity, \(\mathcal{H}_R\) is defined as

$$\mathcal{H}_R = a_w \times 100\%$$.

The relative humidity may also be expressed with reference to the saturation humidity,

$$\mathcal{H}_R = \frac{\mathcal{H}}{\mathcal{H}^*}$$.

The saturation humidity, \(\mathcal{H}^*\), occurs when the gas vapour mixture is in equilibrium with the liquid, i.e., \(p = p_{\text{sat}}\).

Static equilibrium is defined as a set of conditions under which a material does not exchange moisture with its environment. The water activity is defined under equilibrium conditions which, by definition, implies that the partial pressure of water vapour at the surface, \(p_{\text{surface}}\), is equal to the partial pressure of water vapour in the surrounding bulk gas, \(p_{\text{bulk}}\). In non-equilibrium situations, moisture exchange between the product and its surroundings, i.e., drying, is driven by a difference in these partial pressures. Drying may equally well be considered to result from a humidity driving force. The rate of mass transfer from a saturated surface is

$$\frac{dM}{dt} = KA \left(p_{\text{sat}} - p_{\text{bulk}}\right) = kA \left(\mathcal{H}^* - \mathcal{H}_{\text{bulk}}\right)$$,

where \(K\) and \(k\) are mass transfer coefficients based on partial pressure and humidity driving forces respectively. \(M\) is the total mass of the droplet and \(A\) is the area available for mass transfer.

The atmospheric humidity in equilibrium with a given solid moisture content is given by a sorption isotherm. Because of the complexity arising from the numerous mechanisms for moisture uptake, adsorption isotherms for a given material must, in practice, be experimentally determined. The water activity of most hygroscopic products is not very strongly dependent on temperature. The significance of this observation is that the same sorption isotherm may be used to describe the moisture equilibrium in particles across a range of ambient temperatures.

Free or active moisture is that in excess of the equilibrium level. This may consist of unbound and some bound moisture. Only free moisture may be evaporated in a drying operation. The precise drying characteristics of a material depends upon whether it is bound or unbound moisture which is being removed. Whilst unbound moisture remains, the partial pressure of moisture of vapour at the surface is equal to the saturation vapour pressure and drying proceeds at a constant rate. This situation continues so long as the rate of diffusion within the droplet is fast enough to maintain saturated conditions at the surface. Once there is no unbound water at the surface, the moisture vapour pressure adjacent to the drying droplets falls, causing the drying rate to decline. This continues until the equilibrium moisture content is reached, at which point drying stops.
Feed specification as in Table 1:
Product moisture content 8%
Exit air temperature 82°C
Relative humidity at exit 17%

Table 2: Sample process results and the sorption isotherm used to calculate the moisture equilibrium at the outlet.

### 2.2.4 Equilibrium Based Models

An improvement can be made to generic heat and mass balance models, §2.2.2, by making use of the equilibrium idea introduced in the last section. Recall that to close the heat and mass balance equations, it was necessary to input the outlet product moisture content, along with a complete specification of the inlet streams. If instead it is assumed that, at the outlet, particles are in equilibrium with their surroundings, then use of a sorption isotherm enables the outlet product moisture content may be predicted.

**Example**

Recall the previous example, summarised in Table 1. Here it was shown that the suggested drying operation was feasible. It is now sought to predict the product moisture content at the dryer outlet using the equilibrium assumption. For this, a sorption isotherm is required, relating the moisture fraction of the solid to the relative humidity of the surrounding air. This is shown in Table 2. The calculations show that under the feed conditions specified, the product will leave with a wet basis moisture content of 8%, *i.e.*, the 7% figure guessed previously is thermodynamically unattainable. The air leaving the dryer will be at 82°C with a relative humidity of 17%.

An *approach-to-equilibrium* factor may be employed to handle situations where the product outlet stream is not in equilibrium with its surroundings. However, as a user specified parameter, this provides little insight. Hence, the equilibrium models are really only of any use where equilibrium is attained. Intuitively, this is more likely to be valid for small particles with long residence times in the dryer. Ozmen and Langrish (2003) provide experimental evidence that this is indeed the case for particles with a final size of 30 µm.

### 2.2.5 Rate Based Models

When it is not valid to assume equilibrium conditions at the outlet, the rate at which moisture is removed from the sprayed particles must be considered. Alongside this, it is necessary to
model the particle residence time in the drying unit. Together, these allow the product moisture content at the exit to be accurately predicted. However, it is far from trivial to accomplish either of these objectives.

There are two principle ways in which particle residence time in a spray drying tower may be modelled. The first is to invoke simplifying assumptions about particle motion. Such an approach was used by Clement et al. (1991) who modelled the gas in the dryer as a single well-mixed mass. This allowed them to use the standard expression for the residence time distribution (r.t.d.) of a perfectly mixed reactor,

$$E(t, \tau) = \exp\left[-\frac{t - \tau}{\tau_r}\right],$$

where $E(t, \tau)$ gives the fraction of particles which entered the chamber at time $t - \tau$ that are still resident at the time $t$. $\tau_r$ is the mean residence time of particles in the spray drying tower. Coupled to this model for residence times, the authors used a shrinking core model to describe single particle drying. It was further assumed that particle moisture content was purely a function of residence time in the dryer, i.e., for a given feed flow, the atomiser was assumed to produce a population of identical drops.

The strengths of this model include the ability to investigate dynamic changes in inlet conditions and flowrates and relatively fast computational times. Birchal and Passos (2004) use the same physical model to simulate the drying of milk emulsions, although they present an improved solution algorithm. Both papers present results which exhibit fair agreement with experimental milk drying data.

The main weakness of this approach lies in the assumed residence time distribution. Perfect mixing is nothing more than a first approximation which, for many towers, will prove insufficient. As commented in the paper, it is fairly easy to substitute an alternative r.t.d., although this will almost certainly be unique and thus have to be measured for each unit to be modelled. Thus, whilst this approach is useful for investigating changes to operating conditions on existing pieces of kit, it is not an adequate tool for detailed design, (Reay; 1988).

### 2.2.6 Rate Based Models Using CFD

The most sophisticated level of spray dryer modelling is to extend rate based models by introducing a detailed description of the fluid flow within the equipment. This is a non-trivial problem, requiring the detailed solution of a two phase system, with energy and momentum coupling between the phases. There are three fundamentally different approaches to solving two phase flow problems.

It is possible to treat both phases as a continuum, the fully Eulerian or ‘two-fluid’ model. This approach is widely used for predicting two-phase flows with high volume fractions. Platzer and Sommerfeld (2003) demonstrate the use of an Euler/Euler model applied to the dense spray region around a spray nozzle. The method delivers velocities and volume fractions of both phases as output parameters, but no information about the droplet size distribution. This is a major shortcoming of this approach, especially in regions where the spray is dilute and consequently the particle size distribution is very important. Conversely, a purely Lagrangian approach has been demonstrated by Salman and Soteriou (2004). They claim to have identified a fundamental problem concerning volume conservation with the Lagrangian-Eulerian approach when applied to evaporating spray systems. However, provided the systems are sufficiently dilute, this is not likely to prove a major problem.
The vast majority of spray dryer simulations use a combined Lagrangian-Eulerian approach, that is, the gas phase is treated as a continuum, (Eulerian viewpoint) with the droplets being tracked through the flow field in a Lagrangian manner. Such an approach is sensible where one phase occupies a very small fraction of the total volume of the solution domain, (Huang et al.; 2003). The continuous gas phase is modelled using the methods of computational fluid dynamics, (CFD) with the particles being tracked through this flow field. The first work in this area was conducted by Crowe (1980), who used the ‘Particle Source-in-Cell’ method to handle the interphase coupling. This considers the particles as a local source of momentum, kinetic energy and dissipation rate of turbulence. The technique is now standard and built into commercially available CFD packages, such as STAR CD, (Various; 2004).

Fletcher et al. (2003) conducted a review of what factors were important in the simulation of spray dryer performance. They identified four key areas. The first of these is the CFD approach used to model the gas phase. It is reported that, provided a sufficiently fine mesh was employed, the relatively simple Reynolds Averaged Navier-Stokes (RANS) equations were adequate. Furthermore, closure of these equations with the standard \( k-\varepsilon \) model produced satisfactory results. A differential Reynolds stress model has been shown by a number of researchers, (Oakley and Bahu; 1991; Zbicinski and Zietara; 2004), to better predict the flow field where strongly swirling conditions exist, e.g., in the vicinity of the atomiser. The computational effort involved is substantially greater than that associated with the \( k-\varepsilon \) model and, as such, the \( k-\varepsilon \) model is often deemed an acceptable compromise between accuracy and efficiency, (Langrish and Fletcher; 2003). Nevertheless, Bayly et al. (2004) show that such a model does not correctly predict the flow field around the atomiser. Clearly, careful thought is required when selecting a turbulence model.

The remaining three important effects to model are all related to the particulate phase. The most fundamental of these is droplet drying. In its simplest form, a particle drying model will give the rate at which moisture is lost and, from this, the average moisture content. More sophisticated models will give the spatially distributed moisture content, the temperature profile and perhaps even describe the morphological development of the particle. Single particle drying models are discussed in detail in Section 3.

Finally, consideration must be given to the interaction of particles with each other and with the dryer wall. Particle-particle interaction may lead to agglomeration: where two particles collide and stick together. Agglomeration determines the structure and characteristics of many products produced by spray drying. Consequently, several industrial dryers operate in regimes where agglomeration is important, especially those running in counter-current mode, (Oakley; 2004). An example is in the production of powdered milk, where agglomeration provides the required ‘instantisation’ properties. Similarly, in many industrial dryers, accumulation of droplets or particles on the dryer walls is a significant problem and, as such, requires modelling. When particles hit each other or the wall they can, at the simplest level, either bounce or stick. Which behaviour results will depend on a number of factors, but crucially on the surface moisture content of the particle involved. Only wet particles are capable of forming the stable liquid bridges during collision which are necessary for sticking, (Zbicinski and Zietara; 2004). Blei and Sommerfeld (2003) present one of the few examples of an agglomeration model where the collision mechanism is dependent on the material properties of the droplets involved. It is shown that a drying model giving the surface moisture content of the particles is a prerequisite for accurately modelling other particle interaction processes.

The Lagrangian treatment of the particulate phase facilitates the incorporation of physically motivated models to describe the drying and particle interaction processes. Combining sub-models for each of these processes with a CFD simulation of the gas flow would give the basic
structure of a spray dryer model which captured all the important physical processes occurring within such units. Such a scheme is illustrated in Figure 4. However, there are currently very few published results which even attempt to do this.

![Diagram of Full Spray Dryer Model]  
**Figure 4:** Schematic showing the basic structure of modern spray dryer models

A large body of work exists investigating gas flow patterns within spray dryers. Bayly et al. (2004) present a comparison of predicted and experimentally measured flow patterns. The comparison was conducted under cold conditions without any spray present. Huang et al. (2003) use CFD to conduct a detailed investigation of the flow patterns in four different chamber geometries. The authors include water droplets in the flow, but do not consider the detail of the drying of these droplets. A number of authors, (Southwell and Langrish; 2000; Fletcher et al.; 2003; Oakley; 2004) have conducted reviews of the current state of CFD in modelling flows in spray dryers.

Langrish and Fletcher (2003) look forward and conclude that commercially available CFD packages are already capable of producing adequate simulations of the gas phase. They opine that the area in need of most work at present is looking at the adhesion and cohesion of particles due to ‘stickiness’. This is a pre-requisite to producing physically realistic predictions for the rates of these processes. Although ‘stickiness’ is a common concept, its nature is highly complex and it is still poorly understood, (Kudra; 2003).

To date, the only project to attempt to address all the key features found in spray dryers is the ‘Efficient Design and Control of Agglomeration in Spray Drying Machines’ (EDECAD) project, (Verdurmen et al.; 2004). Initiated in 2001, this multinational, EU funded enterprise aimed to develop an industrially validated model to predict agglomeration in spray drying machines, (Verdurmen and Verschueren; 2005). The principle deliverable of the project was a ‘Design-Tool’ to relate the dryer geometry, process conditions, product composition and final powder properties. Those involved with the project claim that the model will be applicable to both co- and counter-current units, although validation has only been conducted with co-current devices.

The particle drying sub-model used is based on the work of Sano and Keey (1982), although this is simplified so that evolving particle morphology is not considered. Particle-particle collisions are treated using the model of Sommerfeld (2001), with agglomeration treated using a classical model of Brazier-Smith et al. (1972). Shortcomings of this approach include: the simplistic drying model; an agglomeration model specifically designed for water droplets; and a structure which does not generalise well to multivariate cases. Furthermore, particle interaction with the dryer wall is not addressed explicitly. It is clear that, whilst considerable progress has
been made towards the development of a unified dryer model, there is room for improvement in all three sub-models.

3 Particle Drying

As a first step towards developing a full, spatially resolved spray dryer model, the decision was taken to first focus on the particle drying sub-model. This section reviews the literature relating to the physics of particle drying and the modelling approaches employed. Section 4 introduces the new work started towards the development and implementation of a new particle drying model.

Drying is a broad and often complicated field. There are many different drying mechanisms, reflecting the compositions, structure and geometry of the particular case being investigated. In a spray dryer, the removal of moisture from a spray involves simultaneous heat and mass transfer. As discussed in Section 2.2.3, the thermodynamic driving force for drying is the difference in water activity between the droplet surface and the bulk. However the drying rate will, in general, be a function of temperature, humidity and the transport properties of the gas and the temperature, diameter and relative velocity of the drop, (Ranz and Marshall; 1952). As the droplets leave the atomiser and contact the drying air, they gain heat from the surroundings by convection. This heat is used to vaporise the moisture, which is then transported to the bulk via the a boundary layer.

3.1 Drying Rate Profile

The drying rate history for an individual particle is shown schematically in Figure 5. The droplet enters the dryer at point A and rapidly heats up until heat transfer with the surroundings reaches equilibrium at point B. Oakley (2004) gives an example showing this period lasts 0.1 s for a 300 µm droplet. The line BC represents the continuation of this condition of dynamic equilibrium. This is the constant rate or first drying period, where moisture removal proceeds at a near constant rate. The surface of the particle, and thus the vapour directly above, remain saturated with moisture and at the wet bulb temperature throughout this period.

The constant rate period will only persist so long as moisture is supplied to the surface at a rate sufficient to maintain saturated conditions. When this is no longer the case, the drying rate begins to decrease, the so called falling rate or second drying period, CD. The transition between these two regimes, point C in the figure, occurs at the critical moisture content.

Although Figure 5 shows a linear fall in drying rate, it is important to recognise that this need not be the case. Local wetted regions may persist at moisture concentrations below the critical value and this will influence the drying rate. Point D represents the concentration at which no wetness remains and the resistance to mass transfer is wholly in the solid layer. The drying rate falls to zero, (E), once the equilibrium moisture content is reached.

3.2 Liquid Droplets

The evaporation of pure liquid droplets has been extensively studied for many years, e.g., Frossling (1938); Ranz and Marshall (1952). Part of the reason why these systems were among the first to be studied is that they are considerably simpler than those involving a higher content of dissolved/suspended solids. Understanding such processes is not only commercially important in itself, e.g., for modelling fuel atomisation in engines, but conclusions drawn from their
Figure 5: A diagrammatic drying rate curve, adapted from Masters (1992)

study also form the basis for understanding more complicated drying mechanisms, (Masters; 1992). For example, Oberman et al. (2004) are working towards modelling the spray drying of colloidal solutions, but first consider the drying of a pure liquid droplet suspended in its own vapour. Interesting conclusions from this particular paper are that the system may be considered isobaric and that surface tension may be safely neglected.

The rate of loss of mass from a pure liquid droplet is given by (6). Rewriting this using a mass transfer coefficient based on mass concentrations, \(k_{ex}\), gives

\[
\dot{m}_{vap} = k_{ex} \frac{W_A}{RT} (p_{\text{surface}} - p_{\text{bulk}}) = k_{ex} \rho_{\text{gas}} (\mathcal{H}^s - \mathcal{H}^{bulk}) ,
\]

where \(\dot{m}_{vap}\) is the evaporative mass flux, \(W_A\) is the molecular mass of the moisture and \(\rho_{\text{gas}}\) is the bulk gas density. Above a pure single component droplet, the air will be saturated, giving \(p_{\text{surface}} = p_{\text{sat}}\) and \(\mathcal{H}^s = \mathcal{H}^*\). A similar expression may be written considering the energy transfer to the particle,

\[
\dot{C}_p M \frac{dT_d}{dt} = hA(T_d - T_{\text{gas}}) + \lambda \frac{dM}{dt} .
\]

Here \(\dot{C}_p\) is the specific heat capacity, \(M\) is the mass, \(h\) is the external heat transfer coefficient and \(\lambda\) is the latent heat capacity of the droplet. Pure liquid droplets will remain at the wet-bulb temperature throughout the drying period. Furthermore, the temperature throughout the droplet may be assumed uniform due to the low Biot number, (Farid; 2003). Under such conditions, the temperature of the droplet is unchanging with time, so (9) can be used to give the shrinkage rate:

\[
\frac{dR}{dt} = \frac{h}{\lambda \rho} (T_d - T_{\text{gas}}) ,
\]

where \(R\) is the current radius and \(\rho\) is the droplet density.

In general, drying models do not consider the spatial distribution of temperature within the
particle. This approximation is considered valid provided that the Biot number is less than 0.1 (Incropera and DeWitt; 2002). The Biot number is defined for a slab as

\[
\text{Bi} = \frac{T_{s,2} - T_{s,1}}{T_\infty - T_{s,2}} = \frac{hL}{k},
\]

and may be thought of as either a temperature ratio, or as the ratio of internal and external mass transfer resistances. Farid (2003) argues that although the constant temperature assumption may be used during the initial drying period, it is certainly not valid throughout drying. He cites the example of dried milk powder where the thermal conductivity of the droplet increases by an order of magnitude as drying proceeds, thus raising the Biot number to values greater than one. This effect is not offset by the particle becoming small. On the other hand, Chen (2004) argues the criteria that the Biot number be less than 0.1 may be relaxed if the effect of surface evaporation is taken into account. Oberman et al. (2004) calculate the internal and external temperature distributions for a pure liquid droplet drying in its own vapour. Their results for a drop with an initial radius of 100 \(\mu\)m show that a uniform temperature distribution, at the wet bulb temperature, is set up within \(\sim 20\) ms. It therefore seems reasonable to assume a uniform particle temperature during the constant drying rate period. However, more care should be taken outside of this regime.

### 3.3 Droplets with Dissolved or Suspended Solids

Particles in real spray dryers will not be pure liquid droplets, rather they will contain dissolved or suspended solids. It is well known that such droplets will evaporate at a lower rate than pure liquid droplets of the same size, (Masters; 1992). Dissolved materials lower the vapour pressure of the evaporating liquid, thus lowering the pressure and temperature driving forces for drying. Annamalai et al. (1993) present a neat analysis of evaporation from two-component droplets, considering the case where both components evaporate. Keey (1992) discusses selective evaporation effects which may occur in such circumstances. Ranz and Marshall (1952) showed that evaporation from droplets with dissolved solids and a single volatile component may always be modelled as if the droplet were saturated with the solute. This is because the solute will crystallise at the drying interface at a rate far greater than it can diffuse away into the centre of the droplet. Conversely, the presence of suspended, \textit{insoluble} solids is found to have a negligible vapour pressure lowering effect and drying during the constant rate period may be treated in the same way as for a pure liquid droplet.

\textbf{Figure 6} illustrates a typical temperature profile during the drying of a particle with dissolved or suspended solids. The temperature is shown to rise slightly in the region \textbf{BC}, implying that the rate of moisture removal is not really constant through the constant rate
drying period. This is true where the liquid has dissolved solids, as the lowered vapour pressure reduces the mass transfer rate, causing the surface temperature to rise above the thermodynamic wet bulb.

Cheong et al. (1986) describe how crust formation will commence when the moisture content falls below at critical value at a single preferential surface site, usually the point of maximum mass transfer as determined by the surrounding flow field. Once initiated, the crust will spread rapidly over the surface of the droplet, forming a structured solid shell and stabilising the particle diameter. The formation of a dried solid layer has a dramatic effect on the subsequent drying behaviour of the particle, marking the end of the constant, and start of the falling drying rate periods. This coincides with point C in Figures 5 and 6. The drying rate is now controlled by the crust resistance to moisture diffusion, where the diffusion coefficient can be orders of magnitude smaller. The drop temperature will rise during the falling rate period, CD, due to reduced evaporative cooling.

Where the ambient gas temperature is greater than the boiling temperature of the moisture in the particle, vaporisation will commence once the particle temperature reaches the moisture boiling point. Considerable energy is required for vaporisation and so the sensible heating of the droplet halts. Dissolved solute raises the boiling point of the moisture and so the boiling point will increase as the solute becomes more concentrated. This explains the gradual rise in particle temperature observed between points D and E. The vaporisation phase is termed the puffing regime and drying rate is now controlled by external heat transfer to the particle, (Hecht; 2004a). A characteristic diffusion length scale may be defined as the average separation between two bubbles. Imagine two adjacent bubbles separated by this characteristic distance, with vapour diffusing radially outwards. If the rate of vapour diffusion from the inner bubble to the outer bubble is smaller than the rate of vaporisation at the inner bubble, then the inner bubble will grow or a new bubble will nucleate between the two. This process will continue until the characteristic length scale has been reduced to the point where the drying rate is equal to the vaporisation rate and the particle temperature remains at the boiling temperature.

Finally, once all the free moisture has been removed, the particle temperature will once again rise, EF, this time until it equals that of the surrounding gas. It is worth noting a high
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initial solids loading will result in the constant rate drying period being brief, if observed at all.

3.4 Drying Models

Although the number of drying models reported in the literature is vast, they are all based on three approaches, (Chen and Xie; 1997). These vary in complexity and are briefly reviewed in this section. Models which attempt to describe morphological development are introduced in the next section.

3.4.1 The Characteristic Drying Curve Approach

This essentially empirical approach has been widely used for the modelling of single particle drying, (Keey; 1992). The methodology is explained in many papers, e.g., Langrish and Kockel (2001); Chen and Lin (2004), and is summarised here. The approach relies upon first identifying an unhindered drying rate which, in this context, means that heat transfer to the surface is limiting, rather than any mass transfer effects. This period may correspond to the rate in the first/constant drying period, where such a regime exists. The relative drying rate, \( f \), is then defined

\[
f(\Phi) = \frac{N}{\hat{N}},
\]

where \( N \) is the drying rate and \( \hat{N} \) is the unhindered drying rate. \( f \) is a function of the characteristic moisture content, defined as

\[
\Phi = \frac{X - X_{eq}}{X_c - X_{eq}},
\]

where \( X \) is the volume-averaged moisture content, \( X_c \) is the critical moisture content (§3.1) and \( X_{eq} \) is the solid moisture content which would be in equilibrium with the surrounding gas, (§2.2.3). Such a definition normalises the drying curve to pass through the point \((1, 1)\) at the critical transition and the point \((0, 0)\) at equilibrium. Figure 7a illustrates some examples of such curves.

The lumped parameter drying rate equation, (8), is then modified using the characteristic drying curve to give

\[
N = f(\Phi) \hat{N} = f k_{ex} \rho_{gas} (H_e - H_{bulk}).
\]

The fundamental assumption of this approach is that the characteristic drying curve for a given material is unique and independent of external drying conditions. Furthermore, it is hypothesised that the drying-rate curves for the same material at different operating conditions will be geometrically similar, i.e., the normalised drying rate curve characterises the drying of a given material. If accepted, this means that drying curves obtained from single particle droplet studies conducted in the lab may be applied to describe drying in industrial scale equipment where the conditions may be significantly different.

Perhaps surprisingly, it is often found that the characteristic drying curve takes on a simple form. For example, Keey (1992) reports that the drying curve can be fitted by a simple algebraic
expression of the form

\[ f = \frac{N}{N_0} = \Phi^n = \left( \frac{X - X_{eq}}{X_{eq} - X_c} \right)^n. \]  

(15)

When moisture movement is limited by the presence of a relatively impermeable outer shell, a first order drying process results and \( n = 1 \) in (15). Conversely, if the drying is limited by the wetted fraction of the exposed surface, then \( n = 2/3 \).

Over the past four decades, there has been considerable investigation into the validity of the characteristic drying curve assumption. Keey and Suzuki (1974) investigated the theoretical foundation of the approach, based on a shrinking core analysis and defining the Kirpichev number, \( Ki \), such that

\[ Ki = \frac{\dot{N}_r}{\rho_S X_0 D_{eff}}, \]  

(16)

Here \( r \) is the thickness of the material, (the radius of a spherical particle), \( \rho_S \) is the density of the dry solid and \( D_{eff} \) is the effective moisture diffusivity through the wet solid. They concluded that the characteristic drying curve may be used when \( Ki < 2 \), i.e., when the material is thin and the effective moisture diffusivity is high. The method might therefore be expected to work for small, micro-porous spheres dried individually.

Keey (1992) provides an overview of the experimental tests of the characteristic drying curve. The results show that the method works well for several materials over a modest range of temperatures, air humidities and velocities. However, the concept did not work for large particles with \( d > 20 \) mm. Also, there is no data reported for drying of slurry droplets.

Recently, Langrish and Kockel (2001) have assessed the suitability of the characteristic drying curve approach for modelling the drying of milk powder. They found that predictions using a linear falling rate curve gave good agreement with experimental data. They point out that the minimal computational expense of this method makes it attractive for use in CFD applications, especially where transient flow patterns are being investigated. However, it is noted that the method gives the volume-averaged moisture content rather real quantity of interest, namely the surface moisture. This point is repeated in the review by Oakley (2004), who concedes that more sophisticated models are required to capture variations in moisture content.

Huang et al. (2004) have specifically investigated the use of characteristic drying curves in CFD models of spray dryers. They used three models, based on the curves shown in Figure 7a. Implementing these models in the commercial CFD package, FLUENT, they demonstrated that the choice of drying model influences both the particle mass history and the path it takes through the spray dryer. Unfortunately, whilst comparing the results from their different drying curves, the authors present no comparison with experimental data.

### 3.4.2 The Reaction Engineering Approach

The reaction engineering approach to modelling of single particle drying was introduced in a paper by Chen and Xie (1997). They claim that this approach maintains the simplicity of implementation associated with the characteristic drying curve (§3.4.1), whilst resting on a more secure physical basis. The method considers drying as a competitive process between an activation type ‘evaporation’ reaction and a ‘condensation’ reaction, (Chen and Xie; 1997; Chen et al.; 2001; Chen and Lin; 2004). That is to say, an energy barrier must be overcome in
order for an evaporation event to occur. Condensation and absorption are not considered to be activation processes. A fractionality coefficient, \( \Psi \), is introduced, which maybe thought of as the relative humidity at the interface between the particle and the drying air. This is expressed

\[
\Psi = \frac{\mathcal{H}^*}{\mathcal{H}^*} = \exp\left(-\frac{\Delta E_v}{RT_d}\right). \tag{17}
\]

where \( \Delta E_v \) is a correction factor for the apparent activation energy for drying due to the increasing difficulty of removing water as the moisture level decreases. When the surface of the particle is saturated with vapour, \( i.e., \) unhindered drying, \( \Delta E_v = 0 \) and the fractionality coefficient, \( \Psi \) is unity. Rearranging gives

\[
\Delta E_v = -RT_d \ln\left(\frac{\mathcal{H}^*}{\mathcal{H}^*}\right), \tag{18}
\]

which provides a means of obtaining \( \Delta E_v \) from drying data. Plotted against the moisture content, \( X \), these curves are considered to be the characteristic property of a particular material under particular ambient drying conditions. For droplets drying under different ambient conditions, it is claimed that there is a characteristic functional relationship between the normalised parameters,

\[
\frac{\Delta E_v}{\Delta E_{v,\infty}} = f_n(X - X_{\infty}), \tag{19}
\]

where \( \Delta E_{v,\infty} \) is the equilibrium activation energy, calculated using \( c.f. \) (18)\)

\[
\Delta E_{v,\infty} = -RT_d \ln\left(\frac{\mathcal{H}^*}{\mathcal{H}^{\text{bulk}}}\right). \tag{20}
\]

The bulk humidity in equilibrium with the particle moisture content at a given temperature may be obtained via the sorption isotherm, §2.2.3. A schematic curve is illustrated in Figure 7b.
Most of the work on the reaction engineering approach to drying is contained within two papers by Chen. In the first paper, Chen and Xie (1997) present a number of case studies where they apply the new approach to modelling the drying of a skim milk droplet, a slab of Kiwifruit leather, silica gel particles and apple and potato slices. More detailed results for the application of the technique to the modelling of the drying of a thin layer of pulped Kiwifruit flesh are presented in Chen et al. (2001). More recently, Chen and Lin (2004) have presented a conference paper in which they apply the reaction engineering approach to the modelling of drying milk droplets. In this paper, they also present a comparison with the predictions of a characteristic drying curve approach. In all cases investigated, the reaction engineering method produces better agreement with experimental results.

Whilst agreement with experimental data is good in all cases, it should be emphasised that a considerable amount of experimental effort is required to obtain both the $\frac{\Delta E_v}{\Delta E_{v,\infty}}$ vs. $(X - X_\infty)$ curve and the desorption isotherm for each system. This drawback is recognised by Chen and Lin (2004). Furthermore, there is still a considerable question as to whether the relationship between ‘activation energy’ for drying and water content is really, as claimed by Chen, a ‘fingerprint’ for the given material. Certainly, it would appear far from clear that this is true when the curve is applied in physical conditions substantially different from those under which it was measured.

### 3.4.3 Models Based on the Diffusion Equation

For droplets of more than one component, or with some solid structure, the moisture and possibly the temperature distribution within the particle will be non-uniform. The most sophisticated drying models employ coupled heat and mass balances to model these distributions. The principle mechanism for moisture movement within such bodies is Fickian diffusion rather than capillary transport, (Efremov; 2002). It is therefore appropriate to use the diffusion equation,

$$\frac{\partial C}{\partial t} = \nabla \cdot (D \nabla C) = \text{div} (D \text{ grad } C).$$

(21)

Although, given boundary conditions, it is possible to solve this equation for an arbitrary 3-D body, simplifications are used in practice, (Li et al.; 2004). Crank (1997) gives many solutions to this equation for different initial conditions and geometries, e.g., for a sphere with fixed external concentration and constant diffusion coefficient, the analytical solution is

$$\frac{C - C_0}{C_s - C_0} = 1 + \frac{2R}{\pi r} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin \left( \frac{\pi n r}{R} \right) \exp \left( -\frac{\pi^2 n^2 D t}{R^2} \right),$$

(22)

where $C_0$ is the spatially uniform initial concentration, $C_s$ is the surface concentration and $R$ is the particle radius. Thermal transport is governed by an equation similar to (21). In spherical polar coordinates, assuming spherical symmetry, this is

$$\frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( k \frac{\partial T}{\partial r} \right),$$

(23)

where $k$ is the thermal conductivity. However, it is noted that the approximation of a uniform particle temperature, §3.2, is often used, e.g., Hecht and King (2000b).
Experimental determination of the diffusion coefficient, thermal conductivity and sorption isotherm are still necessary with this approach. Owing to the need to numerically solve the partial differential equations describing the problem, it is significantly more computationally expensive than either of the previous two methods. However, the advantage is that the method rests on a firm physical basis and gives the spatially resolved moisture content. Droplet sizes in spray drying applications vary between 20 and 500 µm, whereas single droplet drying studies must be conducted on far larger droplets, typically 1-3 mm. The validity of techniques which extrapolate characteristic curves measured for these larger droplets is questionable. For this reason, all of the more advanced models for particle drying are based on solving the diffusion equations.

### 3.5 Models with Morphological Development

For all but single component droplets, the composition and structure will evolve as moisture is removed, §3.3. Walton and Mumford (1999b) identify three distinct categories of particle morphology: crystalline, skin-forming and agglomerate. They assert that each category is evidence of a characteristic drying behaviour. In a later paper, Walton and Mumford (1999a) explain how the same chemical and physical properties which determine the particle morphology may also give insight into particle drying behaviour. For example, the rheological properties of skin-forming materials allow the particles to undergo multiple inflation-collapse cycles. Contrast this with crystalline particles which are less pliable and tend to undergo only partial inflation or form hollow or semi-hollow particles. Tsapis et al. (2005) investigate how physical properties of the drying material may be used to predict the onset of buckling. Their simulations are impressive, but it is currently unrealistic to include this level of detail in a general particle drying model.

More advanced drying models attempt to represent the evolving morphology of a particle during drying. The simplest form of such a model is one that tracks the growth of the porous shell formed when a droplet containing dissolved or suspended solids is dried, §3.3. Cheong et al. (1986) present a receding interface model for slurry droplet drying. The authors show this provides a good match to their experimental drying rate and core temperature measurements. Kadja and Bergeles (2003) use such a receding interface model in their parametric study of the spray drying of coal slurries. However, these models have two principle defects. One of the postulates of the model is that evaporation proceeds at the interface between the dried shell and the wet core. This interface remains at or around the wet bulb temperature, resulting in a large temperature gradient across the crust. Schlunder (2004) observes that this is unphysical and contradicts experimental findings that report no major temperature variations within drying particles. He also claims that such a model violates the laws of multi-phase mass transfer within porous media. To overcome these objections a wet surface model is introduced, which invokes capillary transport through the crust and only allows vaporisation at the particle-air interface. The second problem, as described by Lee and Law (1991), is that they conserve mass only if the porosity of the dried shell is the same as that of the original slurry, a condition which is not generally true. They overcome this by introducing into the model a continuously expanding vapour-saturated space located at the centre of the particle.

The idea of a central bubble had been introduced previously in a pivotal paper by Sano and Keey (1982). Hollow or inflated particles are often observed to be produced in spray drying operations with an example illustrated in Figure 8. Sano and Keey allow a bubble of moisture vapour to form at the centre of the particle when the equilibrium vapour pressure of the moisture within the particle becomes greater than the ambient air pressure. This then expands...
rapidly and inflates the particle. The diffusion equation is solved to track moisture movement through the inflated shell, with evaporation occurring at the particle surface. More recently, Hecht and King (2000b) have used a bubble of inert gas to model the effects of morphological development on drying droplets containing a trace concentration of a volatile compound. The fit with experimental data was shown to be good, (Hecht and King; 2000a). Hecht (2004a) extended this work to simulate slurry drying, but here the central bubble is formed of water vapour. The diffusion equation is solved for a theoretical droplet; a grossly inflated shell with thickness equal to the characteristic diffusion length scale. However, heat transfer to the drop, which is assumed to control the drying rate, is calculated based on a user-defined maximum drop size. The requirement for the inclusion of this arbitrary factor is an unsatisfactory feature of the model.

A novel drying model has been developed by Seydel et al. (2004a). Initially, heat and mass transport is modelled by solving the diffusion equations in a moving coordinate system, chosen such that the number of grid points remains constant as the particle shrinks. Population balance equations are solved at each grid point to describe the formation of solid from a dissolved solute. Once the surface solid concentration has reached a sufficiently high level, a solid crust is formed. The growth of this crust is modelled using a second outer coordinate system. Whilst the inner coordinate system continues to shrink due to moisture removal, the outer coordinate system grows such that the diameter of the particle remains constant. The results of this work are very impressive, with profiles of temperature, solute mass fraction and volume fraction of particles all obtained and compared with experimental data, (Seydel et al.; 2004b). They also show how the internal structure of the particle depends upon the temperature of the drying gas. However, a number of possible extensions are apparent, e.g., it would be good to add a central bubble to simulate inflation.

Finally, it should be noted that all the models discussed in this section require experimental determination of some parameters. Be this a drying curve, or diffusion coefficients it is often the case that the accuracy obtainable for these parameters is what limits the predictive power of the model. It is noted by several authors that the many models published in the literature provide good agreement with the data presented in the same paper. However, it is often the
case that the required parameters are calculated from the same data and hence it is perhaps not so surprising that the fit is good.

4 Work Towards a New Model

Work has started on developing a new particle drying model. As the aim is to incorporate this work into a full spray dryer model, it is necessary that the drying model gives a spatially resolved moisture content. It will also be desirable for the model to describe the evolving particle morphology, as this plays a key role in determining the product properties. It is therefore proposed to develop a model which incorporates the follow three key features:

1. Moisture diffusion: solve the diffusion equations;
2. Solid growth: a population balance;

The decision was taken not to solve for the temperature profile within the particle. Having weighted the evidence presented in the previous section, the low Biot number approximation will be invoked to justify the assumption of a uniform particle temperature.

The first step is to characterise the product to be dried. It is decided to model a system described as a solution with suspended solids. The solution is considered to comprise a single solute and solvent which, in reality, is likely to be an idealisation of the real system. For example, if the system under consideration were a detergent slurry the solute may represent electrolyte with water as the solvent and zeolite and undissolved electrolyte making up the suspended solid component. Spherical symmetry is assumed, so the problem is to a single spatial dimension. The system is illustrated in Figure 9.

4.1 Definitions

A recurring problem when reading papers discussing multicomponent particle drying is a lack of clarity with regards the notation used. In an attempt to avoid that problem, this section introduces the notation used in this work. For brevity, the following subscripts are used to denote quantities related to each of the three components:

\[ A \] Solvent
\[ B \] Solute
\[ D \] Solid.

Following the definitions in Crowe et al. (1998), the volume fraction of the dispersed, (solid) phase is defined as

\[
\varepsilon = \lim_{\delta V \to \delta V^0} \frac{\delta V_D}{\delta V}.
\]

(24)

where \( \delta V^0 \) is the limiting volume that ensures a stationary average. The bulk density of the dispersed phase is defined as the mass of the dispersed phase per unit volume:

\[
\rho_d = \lim_{\delta V \to \delta V^0} \frac{\delta m_D}{\delta V}.
\]

(25)
Figure 9: Schematic representation of the model drying particle. The system consists of a solid phase with volume fraction $\varepsilon$, dispersed within a solution characterized by an effective diffusivity, $D_{\text{eff}}$.

Similarly, the bulk density of the continuous phase is defined

$$\rho_c = \lim_{\delta V \to 0} \frac{\delta m_c}{\delta V},$$

where $m_c = m_A + m_B$ is the mass of the continuous phase. The mixture density is given by the sum of the bulk densities for the dispersed and continuous phases,

$$\rho = \rho_d + \rho_c.$$  \hspace{1cm} (27)

The material, or actual densities of the two phases are given by

$$\rho_d^0 = \rho_D^0 = \frac{m_D}{V_D},$$ \hspace{1cm} (28)

and

$$\rho_c^0 = \lim_{\delta (V_A + V_B) \to 0} \frac{\delta (m_A + m_B)}{\delta (V_A + V_B)}.$$ \hspace{1cm} (29)

Note that, whereas the material density of the discrete phase is constant, the material density of the continuous phase varies with radial coordinate and time. This is because $\rho_c^0$ is a function of the local composition. The mass fraction of the solvent is defined

$$\omega_A = \lim_{\delta (V_A + V_B) \to 0} \frac{\delta m_A}{\delta (V_A + V_B)},$$ \hspace{1cm} (30)

and similarly for the solute. The local concentrations of the solute and solvent are defined with respect to the total volume, i.e.,

$$C_A = \lim_{\delta V \to 0} \frac{\delta m_A}{\delta V} = \rho_c^0 \omega_A (1 - \varepsilon).$$ \hspace{1cm} (31)
and similarly for the solute. It is also useful to define the material densities of each of the components as these are temporally constant:

$$\rho_i^0 = \frac{\text{Mass of component } i}{\text{Volume of component } i} = \frac{m_i}{V_i}.$$  \hspace{1cm} (32)

The following relation can then be derived for the material density of the continuous phase:

$$\rho_c^0 = \rho_B^0 + \frac{C_A}{1 - \varepsilon} \left( 1 - \frac{\rho_B^0}{\rho_A^0} \right),$$  \hspace{1cm} (33)

and the dry mass basis moisture fraction is defined

$$u = \frac{m_A}{m_B + m_D} = \frac{C_A}{C_B + \rho_B^0 \varepsilon}.$$  \hspace{1cm} (34)

4.2 Model Equations

The appropriate partial differential equations for modelling mass transport are derived here. The population balance and equations describing bubble growth have yet to be fully considered.

For a system with spherical symmetry, the diffusion equation written for the solvent gives

$$\frac{\partial C_A}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 n_{Ar} \right) = 0.$$ \hspace{1cm} (35)

Here, $n_{Ar}$ is the mass flux of the solvent relative to stationary coordinates. Following Bird et al. (1960), this may be written

$$n_{Ar} = -D_{\text{eff}} \frac{\partial C_A}{\partial r} + \omega_A (n_{Br} + n_{Ar}).$$ \hspace{1cm} (36)

This equation shows that the diffusion flux relative to stationary coordinates is the resultant of two quantities: $\omega_A (n_{Br} + n_{Ar})$, which is the mass flux of $A$ resulting from the bulk motion of the fluid; and the quantity $j_{Ar} = -D_{\text{eff}} \frac{\partial C_A}{\partial r}$, which is the mass flux of $A$ resulting from the diffusion, assumed Fickian, superimposed on the bulk flow. Note that the diffusion coefficient, $D_{\text{eff}}$, will, in general, be a function of the local composition. If a radial bulk velocity is defined

$$V_r \equiv \frac{n_{Ar} + n_{Br}}{\rho_i^0 (1 - \varepsilon)},$$ \hspace{1cm} (37)

then (36) may be written

$$n_{Ar} = -D_{\text{eff}} \frac{\partial C_A}{\partial r} + V_r C_A,$$ \hspace{1cm} (38)

where (31) has been used to substitute for $\omega_A$. Substitution into (35) gives

$$\frac{\partial C_A}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 D_{\text{eff}} \frac{\partial C_A}{\partial r} \right) - \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 V_r C_A \right).$$ \hspace{1cm} (39)
Considering the solute, the concentration may change due to physical transport, (as for the solvent) or by transport to the solid phase. Ignoring physical transport, it is possible to derive the effect of transport to the solid phase on the solute concentration. Consider a mass balance on the solute and solid:

\[
\frac{\partial m_B}{\partial t} = -\frac{\partial m_D}{\partial t}. \tag{40}
\]

Now, from the definition of \(C_B\),

\[
\frac{\partial}{\partial t} (C_B V) = -\frac{\partial}{\partial t} (\rho_D^0 V_D) = -\frac{\partial}{\partial t} (\rho_D^0 \varepsilon V),
\]
or

\[
V \frac{\partial C_B}{\partial t} + C_B \frac{\partial V}{\partial t} = -V \frac{\partial}{\partial t} (\rho_D^0 \varepsilon) - \rho_D^0 \varepsilon \frac{\partial V}{\partial t}. \tag{41}
\]

If it assumed that there is no volume change on crystallisation, i.e., the density of the solute is equal to that of the solid, then \(\frac{\partial V}{\partial t}\) due to interphase transport is zero and

\[
\frac{\partial C_B}{\partial t} = -\rho_D^0 \frac{\partial \varepsilon}{\partial t}. \tag{42}
\]

Clearly this result relies on the rather strong assumption which should perhaps be reviewed at a later stage. The transport equation for the solute may therefore be written

\[
\frac{\partial C_B}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 n_{Br} \right) + \rho_D^0 \frac{\partial \varepsilon}{\partial t} = 0. \tag{43}
\]

As before, the radial solute flux has two contributions,

\[
n_{Br} = -D_{eff} \frac{\partial C_B}{\partial r} + V_r C_B. \tag{44}
\]

Combining the last two equations gives

\[
\frac{\partial C_B}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 D_{eff} \frac{\partial C_B}{\partial r} \right) - \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 V_r C_B \right) - \rho_D^0 \frac{\partial \varepsilon}{\partial t}. \tag{45}
\]

The convective transport term in the above equations arises from the requirements that the volume in each cell must remain constant and that the fluid is taken as incompressible. The term only cancels if the material density of the solute equals that of the solvent, i.e., the case of equi-mass counter diffusion, \(n_{Ar} + n_{Br} = 0\). For all other cases, \(V_r \neq 0\). Rather, volume conservation requires

\[
\frac{n_{Ar}}{\rho_A^0} + \frac{n_{Br}}{\rho_B^0} = 0
\]

\[
\Rightarrow \frac{n_{Ar}}{\rho_A^0} + \frac{n_{Br}}{\rho_B^0} + \frac{-D_{eff} \frac{\partial C_A}{\partial r} + V_r C_A}{\rho_A^0} + \frac{-D_{eff} \frac{\partial C_B}{\partial r} + V_r C_B}{\rho_B^0} = 0,
\]
where (38) and (44) have been used to substitute for the solvent and solute mass fluxes respectively. Replacing the solvent and solute mass concentrations with mass fractions using (31) gives
\[ V_r \rho_c^0 (1 - \varepsilon) \left( \frac{\omega_A}{\rho_A^0} + \frac{\omega_B}{\rho_B^0} \right) = D_{\text{eff}} \rho_c^0 \left( \frac{1}{\rho_A^0} \frac{\partial}{\partial r} [\omega_A (1 - \varepsilon)] + \frac{1}{\rho_B^0} \frac{\partial}{\partial r} [\omega_B (1 - \varepsilon)] \right), \]
which, after rearrangement and cancelling yields
\[ V_r = \frac{D_{\text{eff}}}{(1 - \varepsilon) (\omega_A \rho_B^0 + \omega_B \rho_A^0)} \left( \rho_B^0 \frac{\partial}{\partial r} [\omega_A (1 - \varepsilon)] + \rho_A^0 \frac{\partial}{\partial r} [\omega_B (1 - \varepsilon)] \right). \] (46)

Now, if this expression is evaluated for a constant solids volume fraction, \( \varepsilon \), and noting that \( \omega_A + \omega_B = 1 \), the following expression is obtained for the bulk radial velocity
\[ V_r = D_{\text{eff}} \frac{\partial \omega_B}{\partial r} \rho_A^0 + \rho_B^0 \left( \omega_A V_R - \frac{\partial C_A}{\partial r} \right). \] (47)

The boundary conditions at the centre are obtained from symmetry considerations,
\[ \frac{\partial C_A}{\partial r} \bigg|_{r=0} = \frac{\partial C_B}{\partial r} \bigg|_{r=0} = 0. \] (48)

The boundary condition at the external surface is more complicated. Let the mass transport flux of solvent vapour away from the particle be denoted \( \dot{m}_{\text{vap}} \). This must then be equal to the mass transport flux of solvent, (38) evaluated at the boundary, \( i.e., \)
\[ \dot{m}_{\text{vap}} = -D_{\text{eff}} \frac{\partial C_A}{\partial r} + V_r C_A \bigg|_{r=R} = -D_{\text{eff}} \frac{\partial C_A}{\partial r} \bigg|_{r=R} + C_A \frac{dR}{dt}. \]
This then gives the boundary condition for the solvent at the external surface of the particle as
\[ \frac{\partial C_A}{\partial r} \bigg|_{r=R} = C_A V_R - \frac{\dot{m}_{\text{vap}}}{D_{\text{eff}}}, \] (49)
where \( V_R = \frac{dR}{dt} \) is the rate of change of particle radius. The boundary condition for the solute at the surface reflects the fact that the solute can not leave via evaporation, \( i.e., \) from (44) the boundary condition is
\[ n_{Br} \bigg|_{r=R} = 0. \] (50)

The rate of evaporation from a drying droplet can be calculated by considering the mass transfer to the surrounding gas, as described in §3.2.
\[ \dot{m}_{\text{vap}} = \rho_{\text{gas}} k_{\text{ex}} (C_{\text{surface}} - C_{\text{gas}}), \] (51)
where \( k_{ex} \) is given by the correlation of Ranz and Marshall (1952),

\[
Sh = \frac{k_{ex}(2R)}{D_{gas}} = 2.0 + 0.6Re^{0.5}Sc^{0.3}. \tag{52}
\]

\( C_{\text{surface}} \) is the concentration of the solvent adjacent to the surface of the particle. This can be related to the solvent content at the surface via the sorption isotherm. The temperature of the drop, \( T_d \), can be calculated from equation (9), where the temperature is assumed uniform throughout.

### 4.3 Numerical Solver

It was decided to solve the model equations using a finite difference scheme with code written by the author for this purpose. Although there are numerous routines for solving PDEs available, it was felt that custom written code would be more flexible. The approach taken is summarised in this section together with some numerical results demonstrating that the method has been successfully implemented.

The diffusion equation is parabolic so must be solved as an initial value problem, **Figure 10a.** As explained in texts on finite differences, (Chung; 2002; Morton and Mayers; 1998) it is possible to write explicit or implicit finite difference schemes to varying degrees of accuracy. The explicit schemes suffer the severe handicap that the mesh ratio, defined

\[
\nu = \frac{\Delta t}{(\Delta r)^2} \tag{53}
\]

must be less than 0.5 for stability. The implicit scheme is unconditionally stable. Taking a weighted average of the two types of scheme yields the so called \( \theta \)-method, deriving its name from the weight parameter, \( \theta \). Taking \( \theta = 0.5 \) gives the famous Crank-Nicolson scheme, with \( \theta = 0.5 \) being the lowest value providing unconditional stability.

The difference molecule for such a scheme is shown in **Figure 10b.** From this it is clear that the solution to the finite difference equations depends on the values at six grid points, both at the current and future time. The equation system which results is tri-diagonal, and can be efficiently solved using standard routines for inverting such matrices. In this work a serial routine taken from Press et al. (1996) was used for this purpose. To test the numerical solver it was applied to the following equation

\[
\frac{\partial C}{\partial t} = \frac{D}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial C}{\partial r} \right), \tag{54}
\]

with boundary conditions

\[
\begin{align*}
\frac{\partial C}{\partial r} &= 0, & r &= 0, & t &> 0 \quad (a) \\
C &= C_s, & r &= R, & t &> 0 \quad (b) \\
C &= C_0, & t &= 0, & 0 &< r < R. \quad (c)
\end{align*}
\]

The first boundary condition arises from symmetry considerations. The second boundary condition describes the case of a constant external concentration, with the final condition prescribing an initially uniform concentration profile within the particle. The analytical solution for this equation may be obtained using Laplace transforms, and is given by (22). Special considera-
tion was required for handling the inner boundary condition. This was achieved using a general technique for handling problems in polar coordinates described in Morton and Mayers (1998). The results are shown in Figure 11a where it is seen that the fit with the analytical solution is near perfect. Indeed, it is actually quicker to obtain the numerical solution as the analytical solution requires the summation of a series that exhibits slow oscillatory convergence.

The model equations are subject to a Neumann boundary condition at the outer surface, \( \frac{\partial C}{\partial r} = \alpha \), \( r = R \), \( t > 0 \) (56).

This problem also has an analytical solution, (Crank; 1997), although it is somewhat more complicated and omitted here. The agreement between this and the numerical solution is confirmed by the results shown in Figure 11b.

Finally, the solver was tested to ensure that it works with a variable diffusion coefficient. The diffusion coefficient was assumed to take the form

\[
D = 3 \times 10^{-8} \left( 1 - \frac{1}{1 + 0.2C} \right),
\]

which is plotted in Figure 12b. It is seen that the diffusion coefficient decreases as the moisture concentration is lowered. The external boundary condition was

\[
\left. \frac{\partial C}{\partial r} \right|_{r=R} = \beta C|_{r=R}, \quad \beta = 2 \times 10^4.
\]

The resulting moisture concentration curves are shown in Figure 12a. Note that in this case no trivial analytical result exists and hence a comparison is not possible.
Figure 11: Comparisons between the analytical solution of (54), solid lines, and those from the finite difference solver, points subject to: (a) Dirichlet boundary conditions with $C_s = 10\text{kmol}^{-1}$; (b) Neumann boundary conditions with $\alpha = 2 \times 10^5$. Both simulations were run with $t_{\text{stop}} = 5\text{ s}$, $D = 3 \times 10^{-8}$, and a mesh ratio, $\nu = 4 \times 10^8\text{ sm}^{-2}$.

Figure 12: (a) Numerical solver test with a variable diffusion coefficient. (b) $D$ as a function of moisture concentration, $C$. The mesh ratio used was, $\nu = 4 \times 10^8\text{ sm}^{-2}$.
4.4 Moving Grids

The present problem is one that involves a moving boundary. During the first drying period the particle physically shrinks as moisture is removed. Once a solid structure has formed, the particle retains the same physical dimensions, but the interface between dry and wet material moves with time. It is therefore necessary to give consideration to the manner in which this moving boundary problem will be handled.

There are a number of different methods for dealing with free and moving boundaries. A detailed discussion may be found in Crank (1996). Analytical solutions are available for some specific cases, although these will be of little use for the present problem. Three principle numerical approaches may be identified. It is possible to solve the equations on a fixed grid. This has the disadvantage that the boundary may lie between grid points at some time steps and inevitably the number of grid points in the solution domain will vary as the boundary moves. This will prove problematic for the present application if a population balance is to be solved at each grid point.

The second approach is to transform the problem by a suitable choice of spatial coordinates, such that the boundary is fixed in the new system. Applied to particle drying problems, this idea is the basis of the solids-fixed approach introduced by der Lijn et al. (1972). This has since been used in several drying models, including that of Hecht and King (2000b). However, this does rely on the the mass of solids remaining constant throughout the drying process, which is not true when solid is being formed by solute crystallisation. For this reason it was chosen not to proceed with this method.

An alternative is to keep the number of space intervals across the entire spatial domain constant at all times. The extent of the spatial domain may vary with time due to processes associated with particle morphological development. If the domain is given by the interval $[0 \leq r \leq R(t)]$ and there are $I$ spatial intervals, then the width of each interval at any time is given by

$$\delta r = \frac{R(t)}{I}.$$  \hspace{1cm} (59)

This representation has the advantage that the domain boundary lies on the $I^{th}$ for all times. Furthermore, keeping the number of grid points constant should facilitate the incorporation of a population balance at a later stage. Figure 13a illustrates the idea, showing how the grid evolves in the spatial domain with advancing time. Note that although the figure shows the grid expanding, there is no reason why the same method should not be used equally well to describe a shrinking domain.

Consider solving the diffusion equation, (21), on such a grid. In one dimension with constant diffusion coefficient this may be written

$$\frac{\partial C}{\partial t} \bigg|_r = D \frac{\partial^2 C}{\partial r^2},$$ \hspace{1cm} (60)

where the left-hand-side gives the temporal evolution of concentration at a fixed spatial location. This may equally well be written following the $i^{th}$ grid line,

$$\frac{\partial C}{\partial t} \bigg|_i = \left. \frac{\partial C}{\partial r} \right|_i \frac{dr}{dt} \bigg|_i + \left. \frac{\partial C}{\partial t} \right|_r.$$ \hspace{1cm} (61)
Figure 13: Grid transformations: (a) a uniformly expanding grid in one spatial dimension; and (b) moving coordinate system, whilst the information rests at the same location in physical space. To satisfy this condition, a 'virtual flux' must be introduced.

Now, the grid moves according to

$$\frac{dr_i}{dt} = \frac{r_i}{R(t)} \frac{dR}{dt},$$

which, on substituting into (60) gives

$$\frac{\partial C}{\partial t} \bigg|_i = \frac{r}{R(t)} \frac{dR}{dt} \frac{\partial C}{\partial r} \bigg|_i + D \frac{\partial^2 C}{\partial r^2}.$$  (63)

It is seen that using a moving grid introduces a virtual radial flux into all equations, the first term on the right-hand-side of equation (63). This allows the the information, i.e., concentration, temperature etc., to rest at the same physical location whilst the coordinate system evolves, Figure 13b. Farid (2003) uses a coordinate transformation such as this to solve for the temperature distribution within a drying particle, i.e., equation (23) is written in the form

$$\frac{\partial T}{\partial t} = \frac{r}{R} \frac{dR}{dt} \frac{\partial T}{\partial r} + \frac{1}{r^2} \frac{\partial}{\partial r} \left( k \frac{\partial T}{\partial r} \right).$$  (64)

The model equations (39) and (45) derived in §4.2 are written in an Eulerian frame. Converted to a Lagrangian frame following a given grid line these read

$$\frac{\partial C_A}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 D_{\text{eff}} \frac{\partial C_A}{\partial r} \right) - \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 V_r C_A \right) + \frac{\partial C_A}{\partial r} \frac{r}{R(t)} \frac{dR}{dt},$$  (65)

and

$$\frac{\partial C_B}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 D_{\text{eff}} \frac{\partial C_B}{\partial r} \right) - \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 V_r C_B \right) - \rho \frac{\partial \varepsilon}{\partial t} + \frac{\partial C_B}{\partial r} \frac{r}{R(t)} \frac{dR}{dt},$$  (66)

respectively. It is seen in this last equation that the solids volume fraction, $\varepsilon$, is required in
order to solve for the the transport of the solute. As (65) and (66) are coupled through the bulk radial velocity, \( V_r \), (47), it follows that the evolving solids volume fraction is also needed to solve (65). Therefore, whilst the functionality of the moving grid has been validated, it is not possible to present any meaningful results until the population balance describing the evolution of the dispersed phase has been incorporated.

5 Conclusions and Further Work

All future work will be orientated towards the project aim of developing a new model of a commercial spray drying tower. Figure 14 gives an overview of the work that is necessary to achieve this aim. It is worth noting how the general scheme of work has been instructed by the envisaged high level structure of the final model illustrated in Figure 4. This makes sense from a strategic viewpoint, as each sub model is quite different in its formulation and implementation. It also makes practical sense to break the overall project down into more manageable pieces which are independently verifiable. The diagram is explained more fully in the remainder of this section.

Figure 14: Schematic illustrating the principal areas of future work towards the development of a full spray dryer model.

5.1 Drying Model

The first task is to finish the development and coding of the drying model. Progress towards this goal is well advanced but, as emphasised in the text, the pivotal importance of the drying model means it is worth taking especial care with this particular aspect of the project. Tasks to complete are the inclusion of a population balance to describe growth of the solid phase and then inclusion of a central bubble. There are a number of potential difficulties with this aspect of the project. Although a self-coded finite difference solver has been proven to work, it may prove sensible to investigate commercially available solution algorithms for the model.
CONCLUSIONS AND FURTHER WORK

partial differential equations. Secondly, it is not possible to properly test the work that has been completed, i.e., the partial differential equations describing moisture transport, until the algorithms updating the solids volume fraction have been incorporated.

Once the drying model has been developed to a stage where meaningful results are obtainable, e.g., particle moisture loss curves, temperature profiles etc., comparison should be made with experimental data. There are numerous sources of experimental single particle drying data, so it should not prove overly challenging to find a suitable system for verification purposes. Nevertheless, it should be noted that numerous parameters such as diffusion coefficients and thermal conductivities, need to be experimentally determined before a new system can be modelled. Furthermore, a description of how these vary with temperature and moisture content is needed. This is likely to be the limiting factor when applying the model to new materials.

Clearly, the drying model under development will be too complicated for inclusion in a CFD simulation without modification. Therefore, a model reduction stage will be an essential final step in the development of this sub-model. Model reduction may take the form of simplification of the physical model, an approach used by Hecht (2004b). However, there are also a number of other promising techniques for achieving the desired model reduction, including response surface methods (Myers and Montgomery; 2002).

5.2 Particle-Wall and Particle-Particle Interaction

Particle-wall interaction is an important physical process that requires modelling. It is also one of the areas where least work has been done. However, there are numerous studies in other areas of chemical engineering looking at fouling, soot deposition in engines etc.. The knowledge learned from these diverse fields could be useful in developing models for the interaction between particles and the wall in a spray dryer. The first step in constructing this sub-model will therefore be to conduct a thorough and wide ranging literature review to get a better idea of the techniques available. It is then likely to be necessary to obtain some experimental data on particle interactions with surfaces. This may be conducted in collaboration with industry. The knowledge gained from these two exercises should guide progress in this area.

As noted in the text, particle-particle interaction, i.e., agglomeration and breakage are important effects in many spray drying towers. Work is being conducted in the group to develop novel particle interaction models for use in CFD, and it is hoped to incorporate these into the final spray dryer model.

It will also be necessary to consider the interaction between the sub-models. This has already been mentioned in relation to understanding how the moisture content of the particles influences their collision behaviour. However, the literature contains little, if any, consideration of how agglomeration in return influences particle drying. That is to say, how is particle drying behaviour influenced by the formation of agglomerates. This is very much an open question which requires addressing.

5.3 Bringing It All Together

The final destination of these sub-models is a spray dryer simulation to be built in a commercial CFD package. The author will need to learn how to use such software, drawing on expertise developed within the research group. Once familiarity has been achieved, the various sub models will be included as user defined functions. Finally, validation with experimental observations from an industrial spray dryer will be attempted.
# 6 Nomenclature

## Upper-case Roman

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>Area available for mass transfer</td>
<td>$m^2$</td>
</tr>
<tr>
<td>$C$</td>
<td>Mass concentration</td>
<td>$kgm^{-3}$</td>
</tr>
<tr>
<td>$D_{eff}$</td>
<td>Effective diffusion coefficient</td>
<td>$m^2s^{-1}$</td>
</tr>
<tr>
<td>$E$</td>
<td>Residence time distribution, eqn (7)</td>
<td>[-]</td>
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<tr>
<td>$\Delta E_v$</td>
<td>Reaction engineering correction factor, eqn (17)</td>
<td>J</td>
</tr>
<tr>
<td>$H$</td>
<td>Humidity</td>
<td>[-]</td>
</tr>
<tr>
<td>$K$</td>
<td>Mass transfer coefficient based on partial pressures</td>
<td>$m^{-1}s$</td>
</tr>
<tr>
<td>$M$</td>
<td>Total particle mass</td>
<td>kg</td>
</tr>
<tr>
<td>$N$</td>
<td>Drying rate</td>
<td>$kgs^{-1}$</td>
</tr>
<tr>
<td>$\dot{N}$</td>
<td>Unhindered drying rate, <em>i.e.</em>, in the absence of internal resistance</td>
<td>$kgs^{-1}$</td>
</tr>
<tr>
<td>$P$</td>
<td>Total pressure</td>
<td>Pa</td>
</tr>
<tr>
<td>$R$</td>
<td>Gas Constant</td>
<td>$Jkmol^{-1}K^{-1}$</td>
</tr>
<tr>
<td>$R$</td>
<td>Particle Radius</td>
<td>m</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
<td>K</td>
</tr>
<tr>
<td>$W$</td>
<td>Relative molecular mass</td>
<td>kgkmol$^{-1}$</td>
</tr>
<tr>
<td>$V$</td>
<td>Volume</td>
<td>$m^3$</td>
</tr>
<tr>
<td>$V_r$</td>
<td>Bulk radial velocity, eqn (37)</td>
<td>$ms^{-1}$</td>
</tr>
<tr>
<td>$X$</td>
<td>Moisture content</td>
<td>[-]</td>
</tr>
</tbody>
</table>

## Lower-case Roman

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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<tbody>
<tr>
<td>$a_w$</td>
<td>Moisture activity</td>
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<tr>
<td>$f$</td>
<td>Relative drying rate, eqn (12)</td>
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<tr>
<td>$h$</td>
<td>External heat transfer coefficient</td>
<td>$Wm^{-2}K^{-1}$</td>
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<tr>
<td>$k$</td>
<td>Mass transfer coefficient based on humidity driving force</td>
<td>$kgm^{-2}s^{-1}$</td>
</tr>
<tr>
<td>$k_{ex}$</td>
<td>Mass transfer coefficient based on concentration driving force</td>
<td>$ms^{-1}$</td>
</tr>
<tr>
<td>$k$</td>
<td>Thermal conductivity</td>
<td>$m^2s^{-1}$</td>
</tr>
<tr>
<td>$m$</td>
<td>mass of a component in a mixture</td>
<td>kg</td>
</tr>
<tr>
<td>$\dot{m}_{vap}$</td>
<td>Rate of evaporative mass flux</td>
<td>$kgm^{-2}s^{-1}$</td>
</tr>
<tr>
<td>$n$</td>
<td>Summation index</td>
<td>[-]</td>
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<tr>
<td>$n_r$</td>
<td>Radial mass flux</td>
<td>$kgm^{-2}s^{-1}$</td>
</tr>
<tr>
<td>$p$</td>
<td>Partial pressure of moisture</td>
<td>Pa</td>
</tr>
<tr>
<td>$p_{surface}$</td>
<td>Vapour pressure adjacent to the particle surface</td>
<td></td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
<td>s</td>
</tr>
<tr>
<td>$u$</td>
<td>Dry mass basis moisture fraction, eqn (34)</td>
<td>[-]</td>
</tr>
</tbody>
</table>

## Greek Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon$</td>
<td>Volume fraction of the discrete phase</td>
<td>$m^3$</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>Latent heat capacity</td>
<td>$Jkg^{-1}$</td>
</tr>
<tr>
<td>$\Psi$</td>
<td>Fractionality coefficient, eqn (17)</td>
<td>[-]</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Density</td>
<td>$kgm^{-3}$</td>
</tr>
<tr>
<td>$\rho^{o}$</td>
<td>Material density</td>
<td>$kgm^{-3}$</td>
</tr>
<tr>
<td>$\tau_T$</td>
<td>Mean residence time</td>
<td>s</td>
</tr>
<tr>
<td>$\Phi$</td>
<td>Characteristic moisture content, eqn (13)</td>
<td>[-]</td>
</tr>
<tr>
<td>$\omega$</td>
<td>Continuous phase mass fraction</td>
<td>[-]</td>
</tr>
</tbody>
</table>
NOMENCLATURE

Subscripts
A referring to the solvent (moisture)
B referring to the solute
bulk referring to the bulk gas
c referring to the critical transition between drying regimes
c referring to the drop
D referring to the discrete phase
eq at equilibrium conditions
R relative to saturation
gas referring to the bulk gas
sat at saturated conditions
w at wet bulb conditions

Superscripts
* At saturated conditions
s Adjacent to the particle surface
sat at saturated conditions
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Cheyne, A., Wilson, I. and Bridgwater, J. (2002). Investigation of structural and processing properties of spray dried detergents. c1 - powder morphology and structure, *Technical Report C1*, The University of Cambridge, Department of Chemical Engineering, Pembroke St., Cambridge. CB2 3RA. CONFIDENTIAL.


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