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978-1-107-01210-3 — Modelling Drying Processes
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1 Introduction

1.1 Practical background

Drying (removing water from wet material) has been a very important processing step for a wide range of human endeavours in our history. The dependence of human society on drying is highly visible. For instance, in food production and preservation, drying is the oldest, most popular and one of the most effective ways to make solid foods and to preserve them as long as practically required. The textile industries need drying processes. Natural fibre-based products such as those from the wood and paper industries also need drying as a critical step in manufacturing. In fact, anything having to do with the particulate products, not just food particles (milk powders, vegetable soup powders and the like) but also detergents, fertilisers, and even paints: drying is critical.

As modern 'material science' industries have started to develop at a speed never seen before in our history, wet chemistry is needed, which requires drying ('dewetting') to form solid products which are more usable and transportable.

Some historical and typical products are shown in Figure 1.1.

Food drying is conducted in many ways. The history of using sunlight to dry fruits goes back thousands of years, dating back to the fourth millennium BC in Mesopotamia (http://en.wikipedia.org/wiki/Dried_fruit). Today, dried fruits have the majority of the original water content removed either naturally, through solar drying or sometimes freeze drying and air drying (with low-humidity air in particular), or 'unnaturally', through the use of specialised dryers or dehydrators powered by electricity or combustion. These unnatural ways include mechanical dewatering, convective air/gas drying, superheated steam drying, electro-osmosic processes, osmotic pressure-driven processes, refractory window drying, freeze drying, vacuum drying, and microwave-aided drying processes, to name a few. If one includes liquid evaporation (with liquid products also), these may be expanded to evaporation operations, such as falling film, rising film evaporation, vacuum distillation, and the like.

Dried fruits are popular products due to their enhanced sweet taste, concentrated nutritional value and long shelf-life because the water activity is low (Chen and Mujumdar, 2008). As water content is removed, the material shrinks (leading to a smaller volume), the sugar content per unit volume of the material increases, as do nutritional components such as proteins, vitamins and so on. Today, dried fruit consumption is widespread. Nearly half the dried fruits sold are raisins, followed in popularity by dates, prunes (dried plums), figs, apricots, peaches, apples and pears (Hui, 2006). Many are

(a) (b)

(c) (d)

Figure 1.1 Some traditional dried products. (a) Broccoli-steam blanched and air dried (kindly provided by Ms Xin Jin, Wageningen University, The Netherlands), (b) air-dried Chinese tea leaves (taken at Xiamen University laboratory), (c) spray dried aqueous herbal extract (particle size is about 80 μm) (taken at Xiamen University laboratory), (d) timber stacked for kiln drying (kindly provided by Professor Shusheng Pang, Canterbury University, New Zealand).

referred to as ‘conventional’ or ‘traditional’ dried fruits: fruits that have been dried in the sun or in heated wind tunnel dryers. Many fruits, such as cranberries, blueberries, cherries, strawberries and mangoes, may be infused with a common sugar (e.g. sucrose syrup) prior to drying to enhance sweetness and microbial stability. This means these are not necessarily healthy products, especially for diabetics. Sugar-infused and dried papaya and pineapples are actually candied fruit. Dried fruits are usually thought to retain most of the nutritional value of the fresh fruits. The specific nutritional content of dried fruits reflects that of their fresh counterparts and is influenced by the processing method or processing technology, particularly processing temperature. In general, all dried fruits provide essential nutrients and an array of healthy protective ingredients, making them valuable tools to both improve diet quality and help reduce the risk of chronic disease.

Furthermore, dried fruits (and nuts) are not only important sources of vitamins, minerals and fibre in the diet but also provide a wide array of bioactive components or phytochemicals. These plant compounds are not designated traditional nutrients since they are not essential to sustain life but play a role in health and longevity and have been linked to a reduction in risk of developing major chronic diseases. Convincing evidence suggests that the benefits of phytochemicals may be even greater than currently understood, as they seem to affect metabolic pathways and cellular reactions. However, the precise mechanisms by which specific compounds exert their biological effect remains largely hypothetical, which requires greater investigation. Certainly, as is well known, dried fruits are an excellent source of polyphenols and phenolic acids (USDA, 2007). These compounds make up the largest group of phytochemicals in the diet and appear to be, at least partially, responsible for the potential benefits associated with the consumption of diets rich in fruits and vegetables. Different dried fruits have unique phenolic profiles (Donovan et al., 1998); for example, the most abundant in raisins are the flavonols (quercetin and kaempferol) and the phenolic acids (caftaric and coutaric acid) (Willamson and Carughi, 2010). Therefore, due to their high polyphenol content, dried fruits are an important source of antioxidants in the diet (Wu et al., 2004; Vinson et al., 2005). Antioxidants can lower oxidative stress and so prevent oxidative damage to critical cellular components. Dried apricots and peaches are also important sources of carotenoids. These compounds not only are precursors of vitamin A but also have antioxidant activity. Dried fruits such as dried plums provide pectin, a soluble fibre that may lower blood cholesterol levels (Tinker et al., 1991). Dried fruits such as raisins are a source of prebiotic compounds in the diet. They contain fructooligosaccharides like inulin, naturally occurring fibrelike carbohydrates that contribute to colon health (Camire and Dougherty, 2003). The chemical structures of some of these useful compounds for human health are shown in Figure 1.2.

Since drying foods may affect the chemical structure of their components, some of which could be undesirable, it is important to keep a balance between how fast or efficient the drying process is in terms of energy usage or product throughput and quality requirements. Sometimes, faster processing is not necessarily better. Like food material, all the solid-form products, natural or processed, have interesting structures (including microstructures) and qualities.

It is worthwhile noting that removing water in its liquid form from a solid structure is often (perhaps better) called ‘dewatering’, which induces solid structural changes around or near where the water used to be. In a more general situation, water removal may also be called ‘dehydration’, especially when talking about materials of partially or wholly biological origin.

In this book, drying is mostly referred as *those processes that use gas as a drying medium so the water comes out of the material as a gas* (water vapour). Later in this book, other forms of drying, such as vacuum drying or even steam-aided drying, may be employed using the *reaction engineering approach* (REA) when it is appropriate.

Furthermore, roasting (coffee, for instance), baking (biscuits and bread, etc.) and heating of moist material (e.g., detoxification of wood) may be considered extensions of the concept of drying, and their purposes are for improving material performance while the moisture effect simply cannot be ignored.

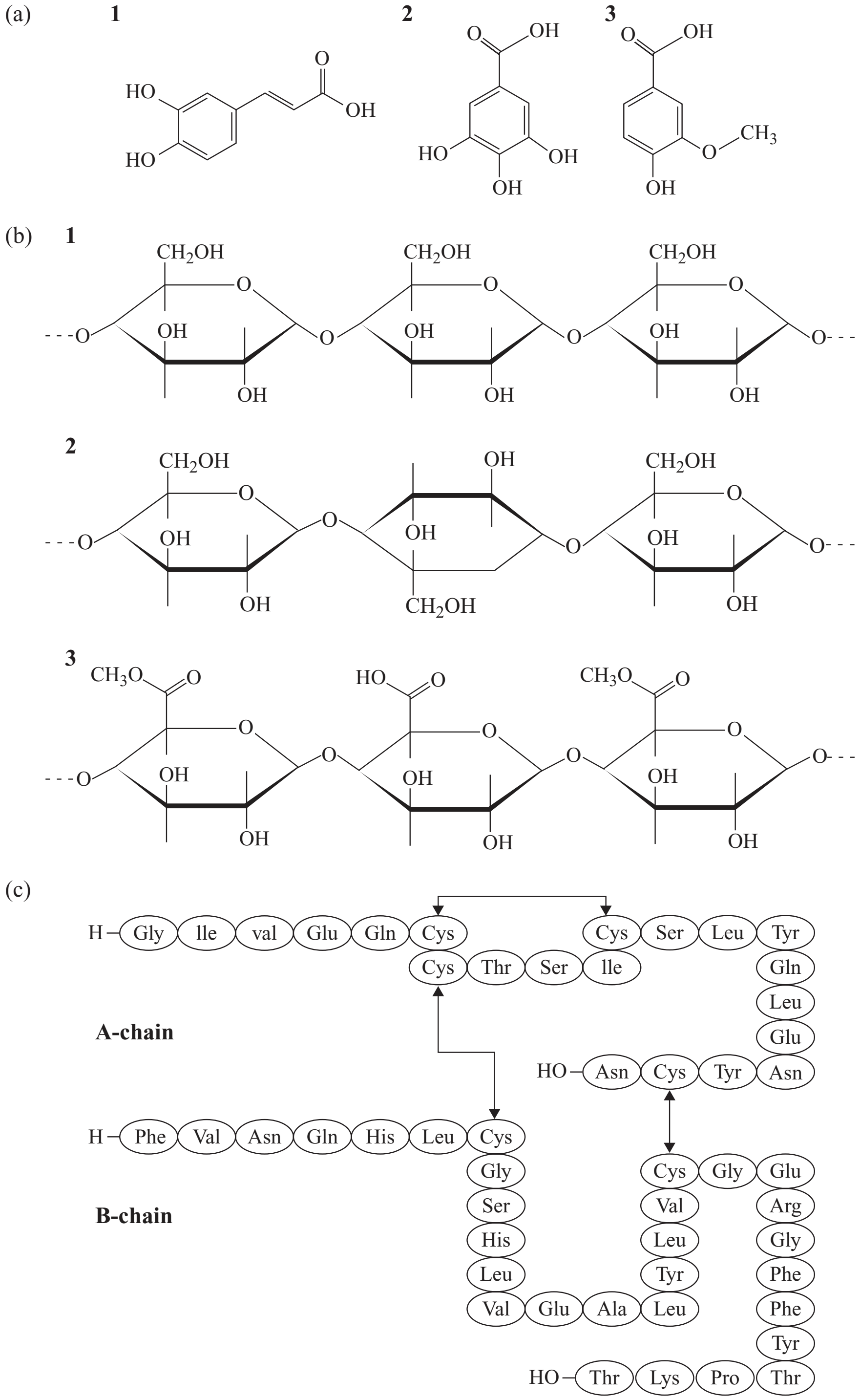

Figure 1.2 Chemical structures of some chemicals: (a) **1**, caffeic acid; **2**, gallic acid; **3**, vanillic acid; (b) **1**, cellulose; **2**, starch; **3**, pectin; (c) human insulin.

The powering mechanisms for drying can be solar radiation, electricity, steam, microwave and ultrasound, amongst others. In all drying operations, energy consumption is a critical issue in modern times; yet, in the wider range of practical interest, people are more concerned about the quality of the products. For high-value products often related to nanotechnology these days, nanostructure and microstructure aspects and functionalities are an increasing concern for both the researchers and marketers. In food and pharmaceutical products, these aspects are even more important as they affect the metabolism and health of our organs.

Therefore, as far as drying is concerned, it provides an excellent example of process engineering interacting with product quality. In chemical engineering, the interactions may be considered systematically with the ideas of *chemical process engineering* versus *chemical product engineering* (Cussler and Mogoridge, 1997).

One may see *engineering* as a terminology which differs from *technology*. Engineering should involve developing the predictive tools (evaluating a building design using established mathematical analysis and then building it accordingly), which need good mathematical descriptions of the physics and chemistry that go on into the relevant processes, much like in modern times, where calculations are used to investigate the validity of constructing an architecturally designed building before actually building it; i.e., civil engineering. There is no question that drying technology needs to be made predictive: this is the notion of ‘drying engineering’. In fact, for design and optimisation of the drying processes, one needs accurate and robust mathematical models. Better still, some of the models can actually help us explore drying mechanisms (physics, chemistry and biochemistry), investigating the scientific aspects associated with the drying phenomena. Drying models are usually referred to as models that describe the mass and heat transport within the material being dried, as the exterior conditions are already nicely covered by the conventional heat and mass transfer and momentum transfer theories. The boundary conditions are intended to connect the drying models with exterior transport theories.

Simple, yet effective, mathematical models of drying are welcomed by practitioners or engineers. When modelling multiphase flow in a spray dryer, for instance, one might need to mathematically track thousands of particles of different sizes travelling inside the drying chamber to make the simulation more realistic. If one has to solve the spatially distributed variables (like water content) inside each particle, the computational effort is great; hence, the whole exercise incurs a high expense. There is also a high likelihood of computational instability. However, if one does not need to solve for the spatial distributions of moisture, temperature and species distribution within each particle, one only needs to integrate a model over time to obtain the average water content, temperature and average concentrations of species for each particle; the computational effort is much smaller.

For a large piece of material being dried, a ‘lumped’ drying model that may predict drying history accurately without the need to resolve the spatially distributed parameters is also very handy for practical purposes. Especially when many such pieces of the same material are placed or stacked in a large chamber to dry (wood stacks for instance), the airflow patterns around these pieces are already complex and need significant

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computational input. The lumped models can be implemented using simple software such as an Excel spread sheet and sometimes even a simple, programmable scientific calculator.

To be relevant to the specific material of concern, every drying model proposed needs careful experimentation to establish the required model parameters (constants) and the model predictive power. In other words, these constants are mostly *material specific*. For a diffusion-based drying model, for example, effective mass diffusivity is the key parameter. It may be both water content and temperature dependent, making accurate experimental determination and data analysis difficult due to non-uniform temperature distribution when it comes to relatively large materials.

If there was one model that incurred only minimal experimental effort and less demand for lab facilities, that model would be welcomed by industry. Relevant experiments have to be accurate, possessing the required resolution, and simple, robust apparatuses and simple operating procedures are desirable.

In many ways, the idea of the REA, apart from its scientific merits and the authors' own desires to make a novel model at that time (1996), is an outcome of these rationales.

1.2 A 'microstructural' discussion of the phenomena of drying moist, porous materials

Drying, the process of water removal, can affect the chemical composition of where the water molecules stayed within the domain marked by the material surfaces before drying. The removal of water molecules leaves vacant spaces, which may be 'filled' partially or totally by a nearby species. These 'movements' should affect the microstructure formation of the material being dried. These movements may include rearrangement of solute molecules (in drying liquid in particular) and shrinkage (with shell formation or hard surface formation as well) and solid structure breakdown (crack formation, for example). Indeed, then a solution (droplet) is dried to form a particle, in a gaseous medium the solid surface formation (chemical composition rearrangement and microstructure formation) is affected significantly by water removal (Chen et al., 2011). The structural changes, once ocurring at nanostructure/microstructural levels, can affect the bioactivity of the biological species, i.e., cells or microbes. The removal of water can cause 'permanent' movement of the soft structures that support the physicochemical structure of the cell wall or cell membranes and cell contents such as genetic material, enzymes, or proteins within, causing irreversible damage. This damage could be minimised if there was another structure-supporting material such as sugar (this can be done by infusing the products, mostly food materials, with sugar molecules in osmotic treatment) which may 'replace' the water molecules to uphold bioactivity. It is also possible that the drying–concentration process alters the ionic conditions, which may be more suited for cells' survival. Drying, in high-temperature instances, with still-considerable moisture content, can cause proteins to denature, thus affecting the solubility and heat stability of protein products. The colour of the food material can be altered during drying, especially when heat is added. Here, Mallard reactions may be responsible where proteins

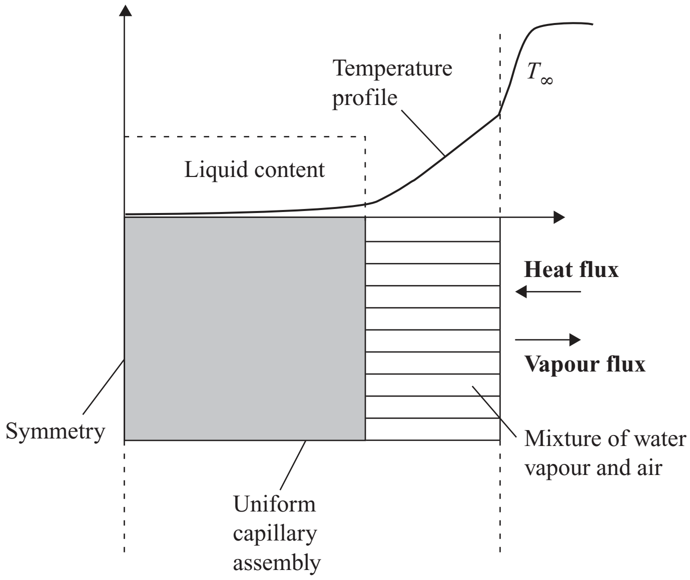

Figure 1.3 ‘Air drying’ of a capillary assembly (a bundle) which consists of identical capillaries (diameter and wall material) – a scenario of symmetrical hot air drying of an infinitely large slab filled with the capillaries (modified from Chen, 2007); the air flows along both sides of the symmetrical material.

and carbohydrates are present, such as in the baking of bread. However, the ‘brown’ colour is welcomed by consumers due to perceptions of a traditional, wholesome and cooked appearance.

Before we proceed with the REA model concept and theory, we first discuss the drying itself in relation to microstructures to provide an important scientific background, which makes drying more relevant to modern material science. *Microstructure* is a relatively new term compared to the classic theories of drying. There are a number of versions of academic descriptions on how drying proceeds into initially moist materials. Here, an intuitive, microstructural view of the gas- (hot air) aided drying process for moist porous media is presented, which is simplified from Chen (2007).

First, we look into a hypothetical scenario and ideal case, where the initial temperature of the moist material is slightly lower than the wet-bulb temperature of the drying medium for an ideal capillary system, as shown in Figure 1.3. The directions of heating and water vapour transfer are opposite each other. The capillaries here have identical diameters at the micro level and the walls are hydrophilic, with no interexchange of heat and mass across the capillary walls (simply, the walls are impermeable), assuming they were initially completely filled with water and evaporation starts to happen. The evaporation occurs uniformly here for all tubes in the same convection condition, at their exits, provided the convection condition is the same everywhere along the side of the assembly. There will be an obvious receding front of the liquid–gas interface moving inwards as drying proceeds. The thickness of the moving evaporation front will reflect the meniscus of the liquid–gas interface.

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A mixture of complexity arises for this ideal system as different diameters of the capillaries and permeable walls (e.g., membranes) are involved. The interexchange of heat is possible across the capillary wall and the evaporation rates among the tubes (under the same drying conditions applied at the exits) now differ. There will be non-uniform receding liquid–gas interfaces among the tubes, giving a distribution of the average liquid–water content (averaged over the lateral direction) along the horizontal direction, broader than that which is shown in Figure 1.3. Furthermore, if the walls are made of materials that are hygroscopic, water-molecular movement or liquid spreading along the wall surfaces is also possible.

Though the interexchange of moisture and heat between the tubes may attempt to even the evaporation rates and liquid water content, a broader distribution of the liquid water content is still expected. Furthermore, due to the extended liquid–gas interfaces, evaporation will not occur only at the meniscus. Evaporation will happen in a region of finite dimension: the occurrence of an evaporation zone at the micro- as well as at the macro-level.

In a non-ideal situation, such as in a normal porous media with pores (open and closed) and interlinking ‘channels’, the likelihood of the occurrence of a sharply receding liquid front is reduced. In other words, the situation shown in Figure 1.3 is *not* common.

When the system is ‘mixed’ at the micro-level (a more realistic situation for a practical porous media), capillaries of various dimensions are oriented in many directions and interlinked or networked. Even locally at the micron level, the capillary diameter sizes can be uneven. The mass transfer is multi-directional, following the laws of physics, that is, following the directions of the driving forces. Locally and microscopically, the receding front(s), if any, would be fuzzy, depending on the specific local microstructure and hydrophilic (or not) nature. Liquid movement may be diffusive or driven by capillary forces and travelling in relatively easier passages. For air and vapour transfer, certain difficult (yet wet) patches may be bypassed by a main ‘receding front’, which may be left to dry more gradually, thus forming a relatively wet region. ‘Fingering’ phenomena may be possible with relatively dry and relatively wet patches coexisting nearby. Here, the sorption/desorption characteristics of the materials distributed should play key roles as well.

The capillary wall’s thickness (the apparently solid structure) and the walls’ own porous microstructures (yet another, smaller level of pore networks or systems), and their unevenness in spatial distribution, add to the overall picture, making the transition more uniform. The materials that create the walls of the microstructures are also important as they can have quite different affinities towards water molecules (these are reflected by their equilibrium isotherms or liquid water holding capacity at the same relative humidity and temperature). Evaporation may occur mostly in the transitional region where the rate is dependent on the local driving force for vapour transfer. The ‘walls’ of the microstructures are also important as they can have quite different affinities towards water molecules (these are reflected by their equilibrium isotherms or liquid water holding capacities at the same relative humidity and temperature).

All these characteristics would make the liquid water content (averaged over these microscopic regions) distributed over a region between the really wet core and boundary

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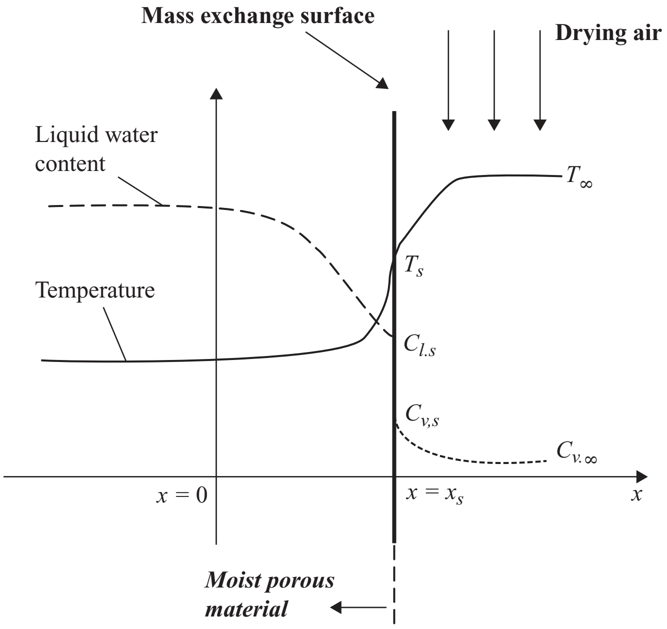

Figure 1.4 Schematic showing a common scenario of air drying of a moist solid.

of the moist material. This leads to a spatial transition of water content distribution rather than the sharp receding liquid waterfront. This is an important recognition of the drying phenomena such as air–gas drying. In freeze drying, on the other hand, sublimation phenomena may induce a sharp solid–gas interface. In high-temperature processes, even for air drying, the powerful heat front may result in an apparently sharper front of water movement than its counterpart when a much lower heat wave is encountered. However, in this case, the previously mentione intuitive analysis is still valid, though quantitatively it may become less influential.

Figure 1.4 shows a general depiction of air drying a moist porous material. One may pay attention to the ‘partitioning’ between the liquid phase and the vapour at the mass exchange surface (the interface between the solid domain and the air). Into the porous solid domain, a vapour phase can also coexist with air and moisture (liquid) phases.

These arguments can be readily generalised to packed particulate systems where the individual particles can have their own macrostructure and sorption characteristics (see Figure 1.5), while the main voids (where easier vapour paths can be found) would be the voids in between the packed particles. In fruits and vegetables, the cellular structure plays a very important role as the cell walls present major water transfer resistance (see Figure 1.6).

The perception of a moving (liquid) front or the (sharp) evaporation front can lead to different approaches in drying modelling. Mass transfer from the sharp moving front and the vapour exit surface is often modelled using a simple effective diffusion concept

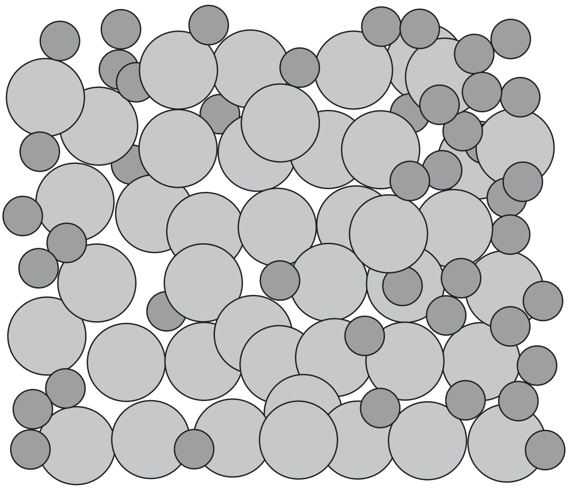

Figure 1.5 Packed particulate material.

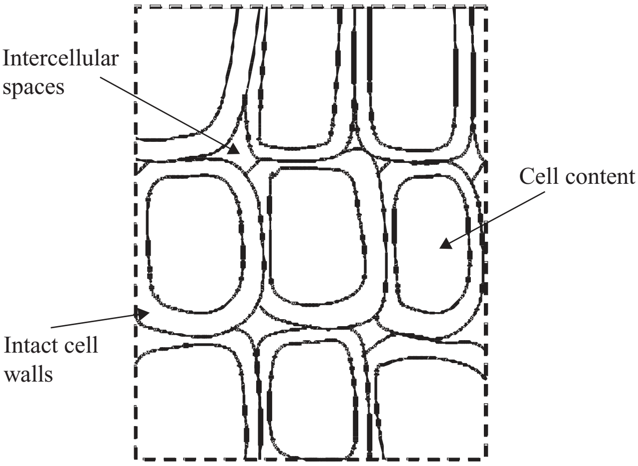

Figure 1.6 Cellular structures in plant material.

(with an expanding resistance layer). In general though, this is a simplification, which is intended largely for mathematical modelling.

It is also interesting to note from the process shown in Figure 1.3 that, due to the temperature distribution in the moist material being dried in a normal air drying situation (where the air temperature is higher than the porous material being dried), it is not necessary to have the highest water vapour content at the innermost boundary where liquid water content starts departing from the initial value. The vapour concentration can be higher than the boundary value, or else there would be little or no drying. It is possible to intuitively reason that there is a ‘hump’ that can exist somewhere in the transitional region of the liquid water content (Chen, 2007). A condensation mechanism also may