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# Modelling the kinetics of drying of d'Agen plums (*Prunus domestica*)

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A two-stage model has been used successfully to predict the drying curves for d'Agen prunes. The model assumes the drying process occurs in two distinct stages. First, a period where the evaporation of water from the surface of the drying fruit is the limiting factor for the moisture loss is assumed. This results in a constant rate of water loss. A second period of drying occurs when a water concentration gradient has been established within the plum. During this time, the rate of moisture loss is limited by the mass transfer of water through the fruit, leading to a falling rate of evaporation of water at the fruit surface. This model predicts reasonable values for the moisture loss as a function of drying time. Significant discrepancies between the experimental results and the model occur only for a longer drying time at the higher temperature range (e.g. 90°C and above). This is due mainly to uncertainty in the value for the equilibrium moisture loss caused by other weight loss processes in the latter stages of drying such as thermal degradation of carbohydrates. © 1997 Elsevier Science Ltd

## INTRODUCTION

Drying food as a means of preservation has a very long history. It is still much used today and its particular appeal is not only due to the extended shelf-life of the dried product by suppression of microbial spoilage. As recently highlighted by Okos *et al.* (1992), other major advantages of dried produce are minimised packaging requirements and lower shipping costs as a result of reduced weight. In some instances the dried (or partially rehydrated) product has it own characteristic flavour and may be thought of as an independent product and different from the original. Such an example is the prune. The prune occupies a small niche market, mainly for the middle-aged and over, because of its mild laxative effect. It is thus sold both as the whole prune, and more recently as prune juice, as a health product. In addition, prune juice is widely used as a flavouring in the food industry in areas such as biscuit making. Prunes are generally produced by drying plums in a dehydration tunnel. The varieties of plums utilised include the Ente, Stanley and d'Agen. All of these are characterised by having a high solids content and usually high sugar levels. (Forni *et al.*, 1992; Newman *et al.*, 1996). The energy expended to dry the plums is significant, constituting perhaps a quarter of the total cost of production. (Bousigon *et al.*, 1988; DFRDC, 1992).

Australia has a small prune industry based in NSW and South Australia. Whilst it is not a major producer, such as France or USA, the annual turnover from the industry is in excess of 4500 tonnes in a good year. The majority of the prunes are consumed domestically (DFRDC, 1992). The Australian industry is under increased competition from overseas producers, in particular South American countries such as Chile, which not only harvest plums at the same time of year as Australia, both being in the southern hemisphere, but have much reduced costs of production (K. Cronk, private communication). The work presented here is part of an industry-sponsored project aimed at investigating the moisture loss process from d'Agen prunes with the intention of identifying ways of increased energy efficiency or revision of drying protocols.

There has been some previous work in the area, notably that of McBean and co-workers in the 1960s, which looked at a number of aspects of the drying process including design of tunnels (counter-flow versus parallel-flow) in both Australia and California. Its main focus was the effect of experimental parameters on final prune quality rather than on the physicochemical basis of the moisture loss process. More recently, there has been a series of investigations by French and Italian

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groups. (Bousigon *et al.*, 1988; Coquinot *et al.*, 1988; Barbanti *et al.*, 1994, 1995) The main thrust in these studies has been the influence of process parameters upon the drying kinetics and on the engineering design of dryers rather than on the physical basis of moisture loss from the plum.

Our previous work on plums has looked at two aspects: a simple first-order kinetic model to describe the drying (Newman *et al.*, 1996) and a study on changes of carbohydrate levels during the drying process. (Wilford *et al.*, 1996, in press) The present paper is aimed at building on the preliminary drying results (Newman *et al.*, 1996) and details the effects of temperature and drying air relative humidity upon the rate of moisture loss. These data are tested against a new two-stage drying model where the drying is characterised by two distinct regimes: a period of constant drying rate followed by a falling one.

## MATERIALS AND METHODS

### Materials

The plums used in this study were d'Agen obtained from the Young district of N.S.W., Australia. The fruits were sorted according to size to obtain an approximately homogeneous sample. Plums weighing between 10 and 20 grams, which represented the average size, were utilised. The samples were kept in sealed plastic bags purged with nitrogen. These were then stored in a refrigerator at 4°C prior to conducting the drying experiments.

### Drying experiments

Prior to the drying experiments, the plums were washed with water and allowed to equilibrate under room conditions. Approximately 1 kg of fruit was used for each experiment. This represented about 50–75 plums. They were uniformly spread on the trays in a single layer and loaded into the drying chamber after the desired drying conditions had stabilised. The placing of the fruit into the oven resulted in an initial decrease in temperature of around 20°C. The oven took approximately five minutes to recover to the drying temperature. However, the subsequent opening of the door during the sampling took less than 1 minute, and it was found that this had a negligible effect on the drying conditions.

A simple forced-air oven (Labec) was used in the study. The unit consisted of a fan, resistance heater and drying chamber equipped with an automatic temperature control device. Inside, the oven had trays for holding the plums. The air-flow was turbulent and was always less than $0.3\,\mathrm{m\,s^{-1}}$.

The weight loss due to drying was monitored at 1 h intervals until the end of the drying period by taking out samples allocated for each time interval. Weighing was carried out using a Mettler P3600 electronic balance which had a sensitivity of ±0.01 g. The drying air temperature and humidity were continuously monitored by using a Vaisala HMP 233 transmitter (Vaisala, Melbourne). The Vaisala probe was factory calibrated but was checked regularly to ensure that it met its specifications. These were: ±0.1°C (in the range −40 to 120°C), and ±1% *RH* absolute (in a range up to 90% *RH*).

Drying experiments were carried out under different conditions of drying air with variables being temperature and humidity. The drying-air temperatures tested were 70, 80, 90 and 100°C. Under the small batch conditions used, it was not possible to accurately control the relative humidity of the air such that it was a constant. In all experiments carried out there was always an initial increase in humidity as water evaporated from the moisture-rich plums. The relative humidity of the drying air was continuously monitored. Three different sets of conditions of relative humidity of the air were achieved by (i) having maximum venting of the exhaust gases, resulting in a LOW value of drying-air %*RH* (0–5%) for the majority of the drying period; (ii), yielding a MEDIUM value of relative humidity (approximately 20–40%*RH*, depending on the temperature); and (iii) restricting the exhaust gases from the drier and introducing a source of moisture into the dryers by way of a tray of heated water placed on the floor of the drier. This resulted in HIGH values of %*RH* (40–65%). Thus, while the control of relative humidity was crude, the system did allow different drying conditions to be sampled. The humidity profiles are shown in the results section and allow the reader to distinguish between the different humidity conditions. The terms low, medium and high humidity will be used throughout the paper to specifically distinguish between the different drying conditions used.

Each drying run lasted for at least 18 h which is consistent with practice in commercial tunnel dryers in Australia. All experiments were repeated at least twice.

Six partially dried plums were removed at each time interval and the weight loss was evaluated. The selection of fruit for removal was carried out in such a way that samples were taken from different sections of the oven to compensate for any small uneven temperature distributions. The oven was tested prior to the drying experiments and temperature variations within the drying chamber were found to be very small (less than 1°C). The dried fruits were then cooled at room temperature before weighing. At the end of the kinetic run for low humidity conditions, samples were also dried continuously until a constant weight was recorded to obtain the experimental equilibrium moisture content for each temperature.

### Measurement of internal fruit temperature

The internal fruit temperature was monitored during drying experiments described above to enable estimates of the average heat-transfer coefficient to be made. This was determined by inserting a copper-constantan thermocouple (0.5 mm diameter) into the flesh of one of the plums adjacent to the stone. The cold junction used was ice (0°C) and the resultant potential difference was measured using a six figure digital multimeter (Hewlett Packard 3468A) and converted to temperature using standard tables (CRC, 1995). The estimated precision of the measured fruit temperature was $\pm 0.1°C$. In preliminary experiments it was established that the difference between the temperature at the centre of the plum and that at its surface was very small for the conditions studied. This was carried out by having two thermocouples, one at the surface and one in the fruit. For example, even during the pre-heating period of the drying, the difference was never more than 2°C at 70°C and diminished to give identical values for virtually all the drying period.

### Initial moisture content

The initial moisture content of the plums was estimated using a standard A.O.A.C. method (A.O.A.C., 1984) for vacuum-drying plums at 70°C for 8 h. This was repeated three times to obtain a reasonable average. The procedure was validated for our equipment by varying the length of drying time to ensure that a constant dry weight had been obtained.

### Mathematical model of prune drying

Many drying processes have been previously studied where the observed kinetics of drying may be divided into two distinct regions, one with a constant rate of drying followed by one which is falling. The falling drying period is one where the rate-determining step is usually mass transfer through the food to the surface under the influence of a concentration and (often) a temperature gradient (Strumillo & Kudra, 1986). The prune is particularly suited to such an analysis as it initially has a very high moisture content. During the first stage of drying there is plenty of water available near the surface and consequently the drying rate might be expected to be a constant, limited by the evaporation rate from the fruit surface. As moisture is depleted, a concentration gradient is created through the fruit. This results in a falling rate of drying. The model used here, therefore, is based on a so-called generalised order kinetics (Chen & Johnson, 1969).

$$(dW/dt) = K(W_e - W)^n \tag{1}$$

where: $(dW/dt)$ = drying rate (kg $h^{-1}$)
$K$ = generalised rate constant ($h^{-1}$)
$W_e$ = final weight loss at equilibrium (kg)
$W$ = weight loss at any time $t$ (kg)
$n$ = order of reaction

The model assumes two distinct periods to describe the total drying curve. The first period is represented by a constant rate of drying. This is immediately followed by a period characterised by a falling rate of moisture loss, given by a single rate constant. Preheating and a second period of falling-rate are neglected. The validity of ignoring the preheating of the fruit was verified by carrying out experiments looking at the fruit temperature profile. Raising the temperature of the plums to a stable value ($T_s$), from an initial fruit temperature $T_i = 22°C$, was a rapid process under the current experimental conditions (taking less than 10–20 mins), and there was no residual thermal gradient within the plum. Utilising a second period of falling-rate is in effect saying that there is a distinct part of the drying process where the diffusion/migration rate of moisture through the fruit is different from that found in the first period of falling-rate. This is neglected in favour of a single average diffusion rate. The model ignores shrinkage of the plum during drying and assumes that no change in the available surface area occurs.

A solution to equation 1 with $n = 0$ (zero order kinetics) was used to evaluate the constant rate period. The fundamental equation in this period has been given by Sokhansanj and Jayas (1987), governing the maximum rate of evaporation from a surface with ample surface moisture.

$$(dW/dt)_c = [hA(T_a - T_s)]/H_v \tag{2}$$

where: $(dW/dt)_c$ = drying rate during constant-rate period (kg $s^{-1}$)
$h$ = heat transfer coefficient (W $m^{-2}$ $K^{-1}$)
$A$ = area of heat transfer ($m^2$)
$T_a$ = dry bulb temperature of drying air ($K$)
$Ts$ = stable fruit temperature during constant period ($K$)
$H_v$ = latent heat of vaporisation of water (J $kg^{-1}$)

This part of the process is limited by the boundary condition ($W = 0$ to $W = W_{cr}$). $W_{cr}$ is the so-called critical weight loss. The critical point occurs when the rate of evaporation starts to become limited by mass transfer of moisture through the plum because of depletion of surface moisture and the setting up of a moisture concentration gradient across the plum. This defines the boundary point between the two periods of drying.

The profile of the falling rate period where the rate of drying is proportional to the moisture concentration gradient can be described by first order kinetics ($n = 1$).

$$(dW/dt)_f = K(W_e - W) \tag{3}$$

At the critical point, which is the junction between the two periods, the generalised rate constant ($K$) in eqn (1) can be calculated from eqn (2) and eqn (3), applying the boundary conditions ($W = W_{cr}$ to $W = W_e$) and assuming a continuous drying process.

$$K = [hA(T_a - T_s)]/[H_v(W_e - W_{cr})] \quad (4)$$

In order to use the above model, two main experimental sets of data are required: first, profiles of the weight loss of the plums with time during drying under particular conditions and, second, corresponding fruit temperature profiles as a function of drying time. The latter allows estimates of the critical drying time ($t_{cr}$) and the stable fruit temperature during the first constant period of drying ($T_s$) to be made by regression analysis. At the critical point it is well known (Strumillio & Kudra, 1986) that the fruit temperature increases from its stable, constant-period value as a moisture depletion layer is formed.

The fruit temperature profiles also enable an estimate of the average heat-transfer coefficient ($h$) to be made. It is assumed that there is no significant temperature gradient within the fruit for most of the constant drying period, and that heat transfer occurs by convection between the air and the plum and by conduction within the fruit. In such a situation the following fundamental equation (Alhamdan *et al.*, 1990) is applicable.

$$hA(T_a - T_s) = MC_p dT/dt \quad (5)$$

where $M$ is the mass of plum and $dT/dt$ is the fruit temperature versus time gradient.

This equation may be solved by integration, applying the boundary conditions ($t=0$, $T=T_i=22°C$; to $T=T_s$ when the fruit temperature has reached a plateau) to yield:

$$[(T - T_s)/(T_a - T_s)] = [(hA)/(MC_p)]t \quad (6)$$

By plotting the experimental data using the above relationship between the initial fruit temperature $T_i$ and the stable fruit temperature ($T_s$) within the constant drying period, an average heat transfer coefficient ($h$) can be evaluated from the slope by regression analysis.

In such an analysis the best available values of the constants in this equation are: $C_p = (1.67 + 2.5\ [IMC]) \times 1000\ \mathrm{J\,kg^{-1}\,K^{-1}}$ (Hallstrom *et al.*, 1988) where $[IMC]$ is the initial moisture content of the plum; and $A = 0.21677\ \mathrm{m^2/kg}$ which was calculated on the basis of the area of an oblate spheroid particle with the following dimensions for a plum: major axis $a = 2$ cm; minor axis $b = 1.5$ cm. (Mohsenin, 1970) This gives a reasonable estimate for the average heat-transfer coefficient ($h$) relevant to the constant drying period.

The determination of $h$ then allows the estimation of the constant drying rate given by equation 2. The best literature estimate (Lydersen, 1983) for the latent heat of vaporisation ($H_v$) for a plum is $H_v = [2501.6\text{-}2.275\ T_s\text{-}0.0018\ T_s^2] \times 1000\ \mathrm{J\,kg^{-1}}$. Having done this, then an estimate for the rate constant $K$ during the second, falling-rate, drying period may be found using eqn (4), and hence a profile for the weight loss during the falling rate period may be determined via eqn (3). To accomplish this a reliable estimate for the expected equilibrium plum moisture loss for given conditions of temperature and relative humidity is needed. This was calculated by using the well-known Henderson equation, (Henderson & Perry, 1955) which is based upon the absolute moisture content of the fruit. The usual form of the Henderson equation is:

$$1 - RH = \exp(-cTM_{eqm}^n) \quad (7)$$

where $RH$ is the relative humidity expressed as a fraction, $M_{eqm}$ is the equilibrium % moisture content of the fruit in units of 100 kg water/kg dry matter, $T$ is the drying temperature in °R (= °F + 459.67), and $c$ and $n$ are empirical constants. The best literature values for plums (Perry & Henderson, 1955) are $c = 1.25 \times 10^{-4}$ and $n = 0.865$. $M_{eqm}$, which is calculated on a dry basis, is related to equilibrium % weight loss $W_e$ via the following simple relation:

$$W_e = [IMC] - M_{eqm}(wb) \quad (8)$$

where $M_{eqm}\ (wb)$ is the equilibrium % moisture content expressed on a wet basis is given by:

$$M_{eqm}(wb) = 100M_{eqm}/(100 + M_{eqm}) \quad (9)$$

## RESULTS AND DISCUSSION

Table 1 details the average weight loss as a function of drying time under six different drying conditions across a range of temperature and relative humidity. The standard deviations for at least two replications were in the range ± 1–7% (typically 2%). As mentioned previously, it was not found possible to maintain a constant humidity, particularly in the first hours of drying. Figure 1 shows typical changes in humidity during the experiments at 80°C under the three different humidity regimes. Initially there is a large increase in the humidity as the water-laden fruit give up their moisture, and subsequently this drops off and reaches a fairly constant value. These plots give the reader an idea of the changing relative humidity conditions sampled. These relative humidity plots are not needed in the model except in the estimation of the final, equilibrium weight loss for each experiment using Henderson's equation. It is the final $RH$% value which will mainly determine the equilibrium moisture content and it is the average final relative humidity % that is calculated from these plots and used in the Henderson equation. The Henderson equation is not, though, very sensitive to errors in the $RH$% value. For example, using the values 90°C and 33% (± 3) relative humidity, one obtains a $W_e$ of 61.5 (± 0.4) %.

The most recent work on prune drying in the literature is that by Barbanti and co-workers (Barbanti *et al.*, 1994, 1995). It is expedient to compare our results with theirs before proceeding further with the analysis. The Barbanti experiments used composite drying conditions with two temperature regimes, but do not quote air humidity conditions. Some of their experiments were conducted on Ente-type plums similar in size to d'Agen. However, using drying conditions of 100°C they obtained a 30% loss in 100 min and a 50% loss in about 200 min. This may be compared with about 110 min and 195 min in the current experiments (see Table 1). This gives confidence that the present results are reasonable and therefore any subsequent analysis is likely to be appropriate. Our preliminary results (Newman *et al.*, 1996) showed similar trends to the present data. For example, in those experiments the temperature dependence was marked, with the initial rate at 70°C being half that at 80°C. This is consistent with the current values. The humidity in the previous experiments was not monitored closely. Newman and co-workers found a 50% moisture loss at 80°C after 6 h drying. This compares favourably with the present results with 53% loss after the same period of drying (see Table 1). The preliminary results have not been incorporated into the present ones because of the uncertainty in the humidity conditions, and because they were carried out using a different batch of plums from a previous growing season.

**Table 1. Mass losses with drying time**

| Drying time (h) | Mean Mass Loss (%) | | | | | |
|---|---|---|---|---|---|---|
| | 70°C | 80°C | 90°C | 100°C | 80°C | 80°C |
| | LOW humidity | | | | MED humidity | HIGH humidity |
| 1 | 5.4 | 9.2 | 14.0 | 19.2 | 6.6 | 6.2 |
| 2 | 11.5 | 20.4 | 25.7 | 33.6 | 13.7 | 10.8 |
| 3 | 18.5 | 29.7 | 37.9 | 47.2 | 20.9 | 15.3 |
| 4 | 25.8 | 39.8 | 47.7 | 54.9 | 27.8 | 20.9 |
| 5 | 32.7 | 48.0 | 52.4 | 62.0 | 33.7 | 24.2 |
| 6 | 37.6 | 53.4 | 58.0 | 66.2 | 39.6 | 27.4 |
| 7 | 44.3 | 59.0 | 61.2 | 69.8 | 45.1 | 32.6 |
| 8 | 48.4 | 61.5 | 65.0 | 71.3 | 49.6 | 35.3 |
| 9 | 52.5 | 64.6 | 67.3 | 72.2 | 53.7 | 40.9 |
| 10 | 55.4 | 66.0 | 68.2 | 73.7 | 57.4 | 42.8 |
| 11 | 58.3 | 67.7 | 69.2 | 74.0 | 59.3 | 46.7 |
| 12 | 60.0 | 68.4 | 70.1 | 74.2 | 61.5 | 48.3 |
| 13 | 61.5 | 69.7 | 71.2 | 74.0 | 61.9 | 51.4 |
| 14 | 63.8 | 69.9 | 71.5 | 74.3 | 63.6 | 52.3 |
| 15 | 64.1 | 70.1 | 72.1 | 74.8 | 64.1 | 54.8 |
| 16 | 64.9 | 70.2 | 72.7 | 75.0 | 65.5 | 56.1 |
| 17 | 64.9 | 71.0 | 72.7 | 75.7 | 66.2 | 57.8 |
| 18 | 65.1 | 71.1 | 73.2 | 75.8 | 66.4 | 58.5 |

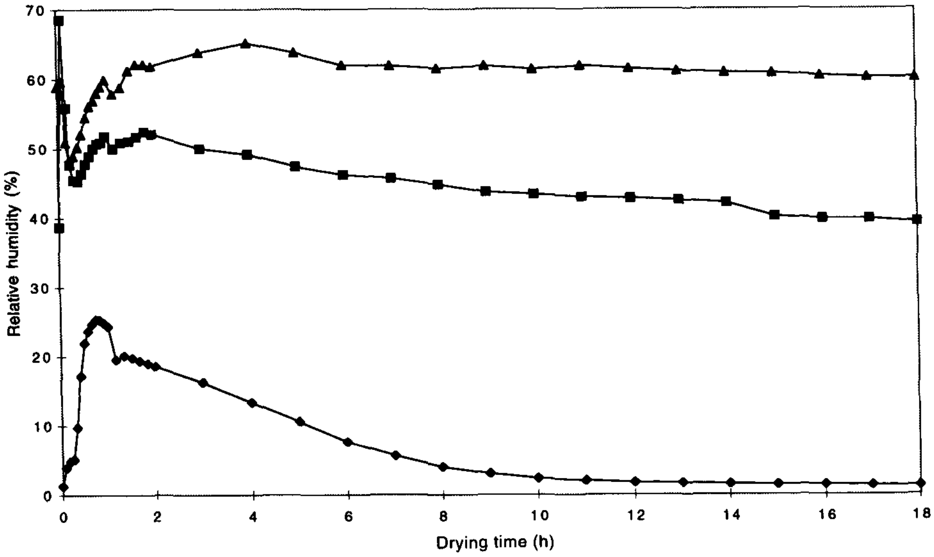

Fig. 1. Relative humidity changes vs drying time for d'Agen plums dried at 80°C under different humidity conditions. Key: ◆ low; ■ medium; ▲ high.

In the previous work (Newman *et al.*, 1996), we used simple first order kinetics (eqn (3)) to describe the complete drying process. This had been used for describing the kinetics of other extraction processes from foodstuffs where the rate limiting step was the transport of the component(s) through the matrix of the food to the surface. This worked reasonably well with successful fits to plots of the integrated form of eqn (3):

$$Kt + a = \ln(W_e/[W_e - W]) \qquad (10)$$

The theory would predict that the constant of integration a should be zero; however, it was found (Newman *et al.*, 1996) that $a$ was always negative. The same is true for the present experiments. For example, using experimental results for 70°C (Table 1), a best line fit of eqn 10 to the experimental data, using an equilibrium moisture loss value of 67.7% (determined using the Henderson eqn), yields a non-zero intercept of $a = -0.35$. The presence of large negative intercepts suggests that diffusion of moisture through the fruit mass is not the only rate-limiting process, but that in the early stages of drying other mechanisms may be important. Figure 2 shows the quality of this comparison between experimental mass loss versus time data for 70°C and data obtained from a best fit of the experimental data to simple first order kinetics. This has been plotted in a mass loss-time form as opposed to the integrated form (eqn 10) to allow the reader direct visual comparison with the new model in later figures. It is seen that the agreement is good at long drying but poorer during the early stages. This is typical of the simple single stage model previously used.

Other work (Wilford *et al.*, 1996) has demonstrated that, particularly in the later drying stages at temperatures of 80°C and above, moisture loss is not the only weight-loss process at play, with chemical reactions including carbohydrates making a significant contribution to the overall weight lost during 'drying'. This often results in the overall weight loss being greater than the initial moisture content. For this reason, and the problems of the old model, particularly in the early stages of drying, it was decided to develop a model that was more physically realistic. The two stage model used here is an attempt to address these problems.

As mentioned previously, many drying processes (Strumillo & Kudra, 1986), particularly those from matrices with a high initial moisture content, exhibit two distinct stages of drying. In order to determine a number of parameters needed in the model, measurements of fruit temperature versus drying time were made. Figures 3 and 4 show results for the temperature profile of the plum during drying for two sets of different drying regimes. These plots are instructive as they typically have two distinct features. In all cases the increase in fruit temperature is rapid, reaching a near plateau region by 20–30 mins. At the higher temperatures this process is even shorter, for example at 90°C it is about 10 minutes. Secondly all the profiles exhibit classical behaviour (Strumillo & Kudra, 1986) in that the temperature after the plateau subsequently shows a small but definite increase. (Admittedly the plateau region is less clear at high temperatures). However, this increase gives an indication of a critical point and evidence that a two regime drying process is taking place. For example, in Fig. 4 the critical point at 70°C (low humidity) is at 5 (±0.4) h whilst at 80 and 90°C it occurs earlier at 3.75 (±0.25) and 2.9 (±0.1) h respectively. These calculations were carried out by linear regression of portions of the data and determination of intersection of those lines to estimate the point where the temperature rises beyond a plateau. This gives some confidence that a two stage model is reasonable in the case of drying plums and that it is possible to obtain estimates of some of the parameters needed from fruit temperature profiles during drying. This will be discussed further later.

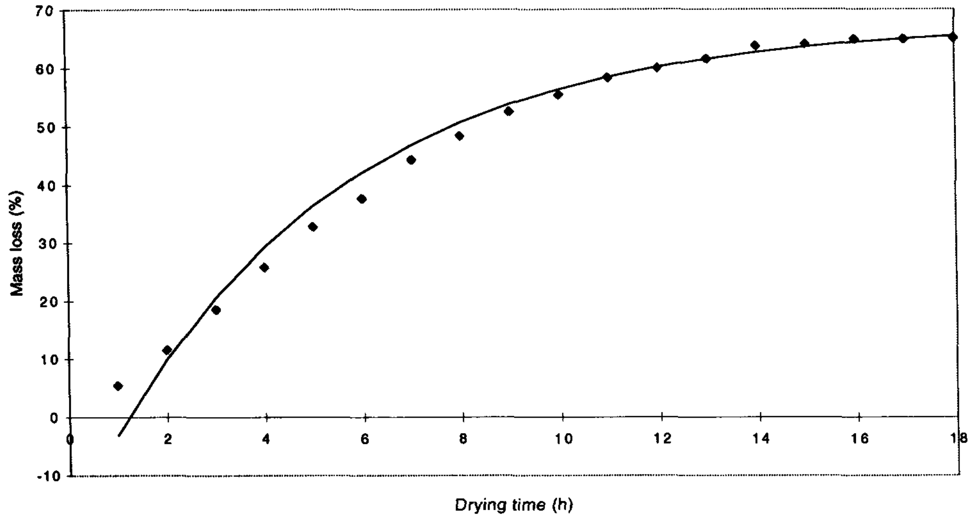

**Fig. 2.** Drying data for d'Agen plums at 70°C fitted to Newman model.

Figures 5 and 6 show the results for the effect of temperature upon the drying kinetics. In Fig. 5 the weight loss versus time curves clearly show the large effect of utilising a higher temperature. The initial rate of drying at 100°C is over twice that at 70°C. Prunes are often dried to about 50% weight loss commercially (representing a residual moisture content of about 18% db). This difference in rate would lead to a change in apparent drying time (under good extraction conditions with the current system) from 12 h to just under 5 h at the higher temperature. However, it should be borne in mind that, apart from the increased fuel costs, another hazard in drying at such temperatures can be higher rates of fruit splitting and consequent loss of contents and hence quality. The other prominent feature of these curves is that the weight losses level out at different values for each temperature, increasing from about 67% at 70°C to 76% at 100°C. The average initial moisture content (*IMC*) of the plums was found to be 67.9 (±1.1) % (on a wet weight basis including the seed). This value was determined from three replications. This clearly indicates that, at higher temperatures, other chemical changes occurring are starting to become significant. These include thermal degradation and caramelisation of the carbohydrates in the fruit. We have previously shown (Wilford *et al.*, 1996) that, at 80°C and above, changes in the simple dominant carbohydrates, glucose, fructose and sorbitol indicate that both Maillard and caramelisation reactions occur in the later stages of drying. This is verified by visual inspection in

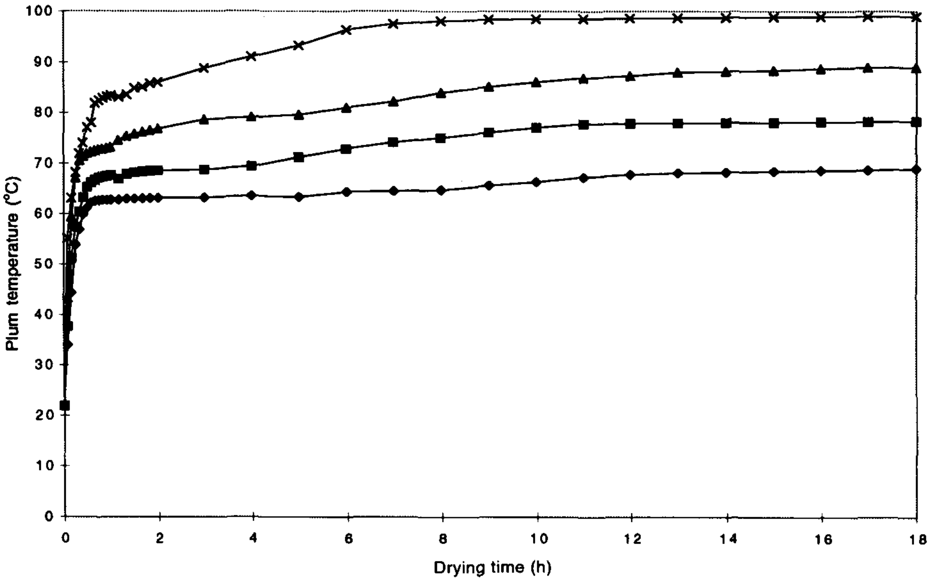

**Fig. 3.** Fruit temperature profile during the drying of d'Agen plums for four different temperatures and low humidity (good exhaust) conditions. Key: ◆ 70°C; ■ 80°C; ▲ 90°C; × 100°C.

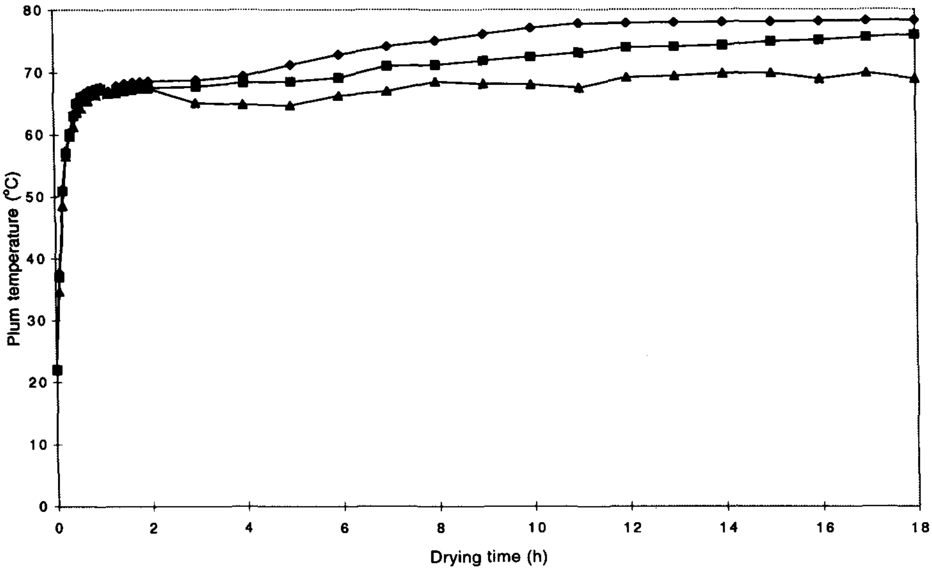

**Fig. 4.** Fruit temperature profile during the drying of d'Agen plums at 80°C for different humidity conditions. Key: ◆ low; ■ medium; ▲ high.

that case-hardening of the dried fruit was evident at the highest temperatures. Rather than ignore the fact that some of the weight loss is due to chemical reactions rather than evaporation of water, it was decided to estimate the equilibrium moisture loss for each set of conditions using an empirical equation (Henderson & Perry, 1955). This allows both analysis of how well the new model fits the experimental data, particularly at lower temperatures, and also, from deviations from the predicted values at the latter stages of drying, an idea of the contribution of other weight loss processes such as caramelisation to the overall weight loss profile.

Figure 6 replots the data for the four different drying temperatures, under low humidity conditions, in terms of the instantaneous rate of drying vs drying time. These plots were determined by calculating a polynomial function for the drying curve, if necessary dividing the curve into two or more parts, and calculating the instantaneous rate from the derivative. The points shown are the average rates calculated at convenient hourly intervals and the error bars show the standard deviation of the rates from the individual experiments. Particularly at the lower temperatures, there are clearly two regimes to the drying, ignoring the initial hour where pre-heating and equilibration are taking place. At 100°C, the rate of moisture loss is very rapid and the constant rate is not apparent with the falling rate decreasing from a very high initial value. This can merely be attributed to the fact that the drying is so vigorous that a water depletion layer is formed very quickly. Investigations into drying kinetics of other foodstuffs have found that, while a two-regime drying is common, the falling rate is the dominant process for most of the drying, such that mass transfer through the

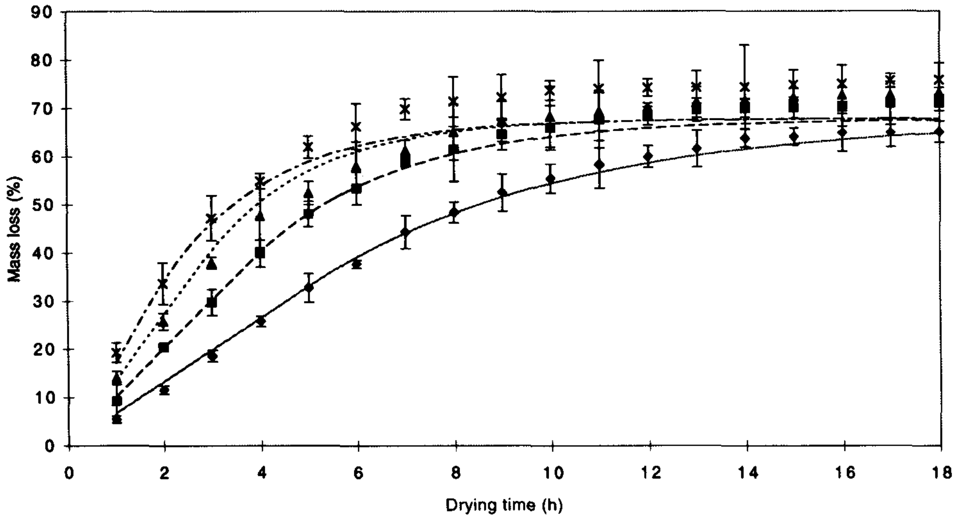

**Fig. 5.** Effect of temperature upon moisture loss from drying d'Agen plums together with predicted loss curves using two-stage model. These are for low humidity conditions. Key: experimental: ◆ low; ■ medium; ▲ high. model: ——— 70°C; — — — 80°C; - - - 90°C; — - — - 100°C

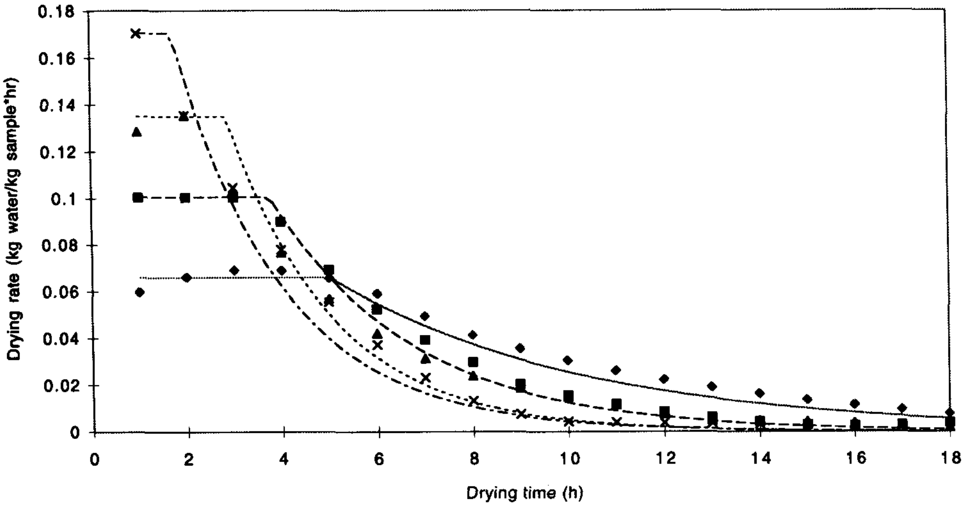

**Fig. 6.** Rate of drying of d'Agen plums vs drying time for four temperatures from 70-100°C for low humidity conditions, and predicted results using the two-stage model. Key: ◆ low; ■ medium; ▲ high. model: ——— 70°C; — — — 80°C; - - - 90°C; — - — - 100°C

fruit to the surface is the rate limiting factor. For a food with a very high moisture content like a plum, a constant drying period might initially be expected due to the large amounts of surface water available.

In a parallel study using pulsed NMR techniques, the mobility of water within the plum tissue is being studied by measurements of the self-diffusion coefficient of water as a function of moisture content. Preliminary results indicate a high initial water mobility with little evidence for any localised restricted diffusion. This is consistent with the observed initial rapid moisture loss during drying.

It is interesting to compare the experimental results for the temperature effect on the drying with the proposed model. The estimates of the drying parameters, such as the average heat-transfer coefficient, determined from the fruit temperature plots, are summarised in Table 2. The values of $T_s$, the stable fruit temperature during the constant drying rate period, and $t_{cr}$, the critical point, were determined by regression analysis and are shown with the standard deviations from the analysis of replicate experiments. The quantities derived from the experimental data using the equations, $K$ and $h$ are given together with the estimated combined error. Values of h compare favourably with those found in other drying systems (Paulus, 1984; Gekas, 1992). The model drying curves are also shown in Figs 5 and 6. A good fit to the drying data at 70°C is found over the entire drying period. For the higher temperatures, the experimental weight loss curves coincide (within experimental error) with the model for the first 5–12 h, when the experimental weight loss starts to lead the model. At longer times, the weight loss at these temperatures exceeds the initial moisture content, as mentioned previously, with consequent deviation from the model.

It may be concluded that, under these low humidity conditions, the two stage model fits the drying data at 70°C extremely well, and reasonably well, at higher temperatures and would be a useful predictive tool for these conditions. In addition, the new two-stage model is superior to the previous first-order kinetic model (Newman *et al.*, 1996), giving much improved agreement with the experimental weight-loss data, particularly in the early stages of drying when the rate of moisture loss is greatest.

**Table 2. Parameters of drying effect at different temperatures**

| Conditions | 70° | 80°C | 90°C | 100°C | 80°C | 80°C |
|---|---|---|---|---|---|---|
| Parameters | LOW humidity | | | | MED humidity | HIGH humidity |
| $T_s$ (°C) | 62.4 (±0.5) | 68.6 (±0.1) | 74.9 (±0.1) | 81.1 (±0.4) | 67.8 (±0.5) | 66.5 (±1.0) |
| $W_{cr}$ (%) | 33.2 (±1.1) | 37.7 (±2.2) | 38.6 (±0.6) | 29.5 (±3.2) | 41.7 (±0.6) | 36.8 (±5.0) |
| $t_{cr}$ (h) | 5.0 (±0.4) | 3.8 (±0.2) | 2.9 (±0.1) | 1.7 (±0.15) | 6.1 (±0.5) | 7.1 (±1.0) |
| $W_e$ (%) | 67.7 | 67.8 | 67.8 | 67.8 | 60.2 | 53.4 |
| $h$ (W/m$^2$ K) | 26.3 (±1.2) | 26.4 (±0.1) | 26.5 (±0.1) | 26.6 (±1.1) | 17.0 (±1.0) | 11.5 (±0.09) |
| $K$ (s$^{-1}$) | 0.19 (±0.01) | 0.34 (±0.04) | 0.46 (±0.01) | 0.44 (±0.05) | 0.38 (±0.14) | 0.31 (±0.14) |

Notes: (1) $W_e$ was calculated using the Henderson equation using the end relative humidity value (averaged) taken from the relative humidity vs drying time plots (Figs 1 and 3)

(2) Uncertainties in $W_{cr}$ and $t_{cr}$ are the standard deviations from the analysis of replicate experiments

(3) Error bars for $K$ and $h$ given are standard deviations of the mean values from combination of errors analysis (see text)

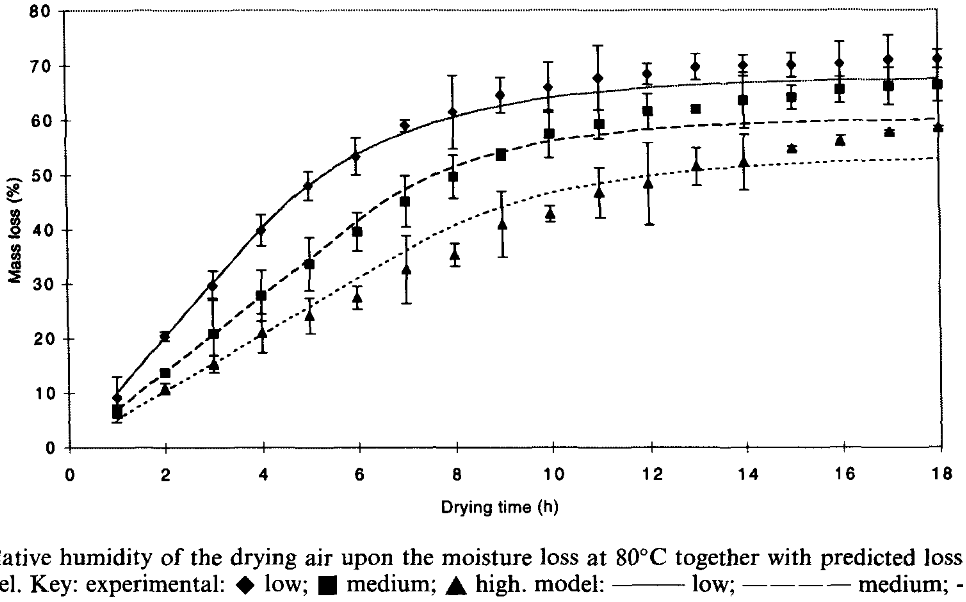

**Fig. 7.** Effect of relative humidity of the drying air upon the moisture loss at 80°C together with predicted loss curves using two-stage model. Key: experimental: ◆ low; ■ medium; ▲ high. model: ——— low; — — — medium; ---- high.

At higher temperatures, it may be seen from the deviations between the model and experiment at longer drying times, that there are other non-moisture loss processes contributing to the experimentally observed weight loss. These may be reasonably ascribed to chemical reactions occurring such that the weight loss is not entirely due to water movement. At 100°C after 18 h, some 10% of the total weight lost is due to chemical reactions. In addition, inspection of all the drying curves at the higher temperatures, and comparison with the predicted model curves, shows that other weight loss reactions, such as Maillard and caramelisation, start to occur quite early on in the drying process. At 100°C, the systematic deviation between the two curves begins at about 6–7 h. This is consistent with previous work studying the changes in carbohydrate content in plums with drying, (Wilford *et al.*, 1996) where decreases in the major monosaccharides glucose and fructose begin after about 6–8 h at 100°C.

Comparison of the drying data at 80°C under different humidity conditions, to ascertain the effect of moisture in the drying air upon the rate of water removal, is shown in Figs 7 and 8, together with the results of the model drying. The higher humidity conditions are seen to have a dramatic effect upon the initial drying rate. The rate of drying, initially at the highest humidity, is less than half that for the low *RH*% conditions. In terms of expected drying time, this would lead to a difference of over 14 h (22 compared to about 8) in reaching a final moisture content of 9–13%. This is a

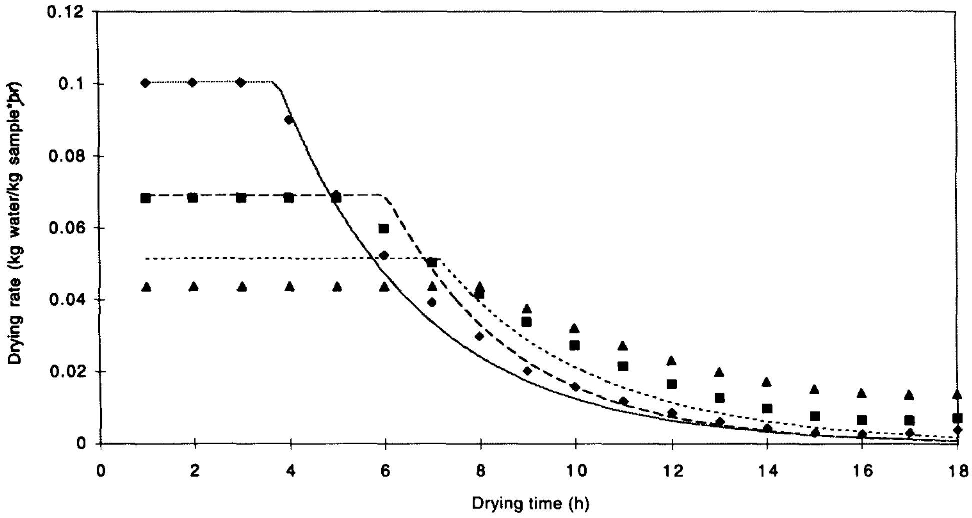

**Fig. 8.** Effect of relative humidity of the drying air upon the rate of drying at 80°C together with predicted rate of drying curves using two-stage model. Key: ◆ low; ■ medium; ▲ high. model: ——— low; — — — medium; - - - - high.

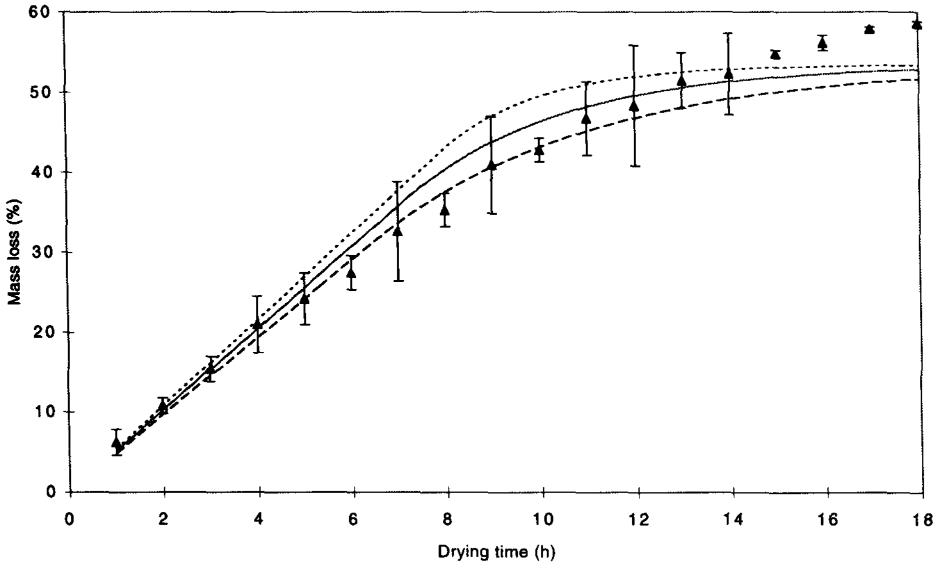

**Fig. 9.** Experimental mass loss curve vs drying time at 80°C and high humidity conditions (end final humidity 60%) together with the model results. Three curves for the model are shown giving the results using the mean of the estimated parameters and the extreme ranges using the standard deviation of the parameters from the analysis. Key: ▲ expt; ——— predicted (mean values); — — — — and - - - - predicted (limits).

dramatic difference and illustrates the importance of monitoring and controlling (if possible) the drying air humidity during commercial drying of fruit.

The fits of the model to these curves in Fig. 7 and 8 are reasonable, all exhibiting the two drying-stage pattern. The problem of other weight loss reactions during the latter stages causes some systematic deviations. However, these conditions in general exhibit a good agreement between the experimental point and the model curve for the majority of the drying time, discrepancies becoming measurable at later drying times. The point at which the deviation between the model curves and the experimental data become manifest is dependent upon the humidity of the drying air. This suggests that the additional mass loss due to degradation reactions is influenced by the moisture content in the drying fruit and not just a function of temperature. This is verified by other work on caramelisation reactions, which has shown that, in general temperatures of 120°C and above are needed to caramelise sugar solutions but that the extent of caramelisation is crucially dependent on water content (Kroh, 1994).

In order to assess the uncertainty in the model, Fig. 9 shows the experimental % mass loss vs drying time at 80 °C and medium humidity conditions together with the model results. Three different lines are shown for the model, the mean and the extreme range of values based upon the uncertainties in the estimated quantities as shown in Table 2. This particular set of data, was chosen because it has the largest uncertainties in $h$ and $t_{cr}$. However, it is clear that even in this case, the model and the experimental results agree well within the combined uncertainty up to perhaps 14 h.

It is also worth noting that, although the Henderson equation to calculate $W_e$ is insensitive to uncertainties in both temperature and relative humidity, there could be a systematic error in the estimated $W_e$ values. This could be a contributing factor to the observed deviation between the experimental results and the model at long drying times. The parameters used in the Henderson equation may not be ideal for the particular type of plum used in this study. This matter, and alternative estimates for the expected equilibrium moisture loss under different drying conditions, are currently being investigated.

## CONCLUSIONS

The proposed two-phase model successfully predicts the drying curves for prunes at temperatures between 70 and 100°C for a range of humidity conditions. The deviations at higher temperatures are seen to be due to other (non-moisture) mass loss processes, contributing to the overall process. Comparison with the predicted curves is interesting as it shows when the other degradation processes start to become manifest. For example, at 90°C (and low *RH*%) this occurs at 9–10 h. This is consistent with previous work monitoring carbohydrate change within drying plums (Wilford *et al.*, 1996). This is a useful way of seeing the contribution of other processes to the drying and hence to the quality of the final product.

The new model is clearly an improvement on the Newman single stage, first-order kinetic model (Newman *et al.*, 1996), giving much better agreement with experiment, particularly in the early vigorous stages of drying. Consequently, the addition of an initial constant rate of drying stage when there is plenty of moisture available at or near the surface is seen to be successful and physically realistic. The model is simple. It relies on only a few parameters and, despite the uncertainties in some of the derived quantities, the model agrees well with the data within the combined error.

We are currently carrying out experiments in a drying system simulating commercial prune tunnel drier conditions. The drying conditions are different in that high velocity (1–5 m s$^{-1}$) laminar air flow is used. It will be interesting to further test and develop the new model under these different experimental conditions.

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