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Diffusive Drying Kinetics in Wheat, Part 2: applying the Simplified Analytical Solution to Experimental Data

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Reanalysis of the drying background in wheat showed that analytical solutions may be employed in this grain to estimate diffusion coefficients by using the simplified equation for short times instead of the time-consuming series. Sixteen thin-layer drying curves of hard wheat were measured (airflow $\approx 0{\cdot}3$ kg m^{-2} s^{-1}) covering four air temperatures (35–70°C) at each of four initial moisture content levels (0·189–0·269 decimal, d.b.). Experimental curves of the moisture ratio versus time grouped by initial moisture content showed the expected strong accelerating effect of temperature on drying rate. Besides, when the same curves were grouped by temperature, the moisture ratios corresponding to higher initial moisture contents fell, after some time, consistently faster, showing that the diffusion coefficient should increase somehow with water concentration. The short time simplified diffusive equation fitted each curve very well, with values of the coefficient of determination above 0·99. Values of the diffusion coefficient for the whole kernel ranged from $1{\cdot}4\times10^{-11}$ to $7{\cdot}1\times10^{-11}$ m^{-2} s^{-1}, presenting the classical Arrhenius temperature dependency (activation energy $\approx 27{\cdot}0$ kJ mol^{-1}), but with a pre-exponential factor that depends linearly on initial moisture content. This diffusive kinetics is expected to be useful for fast and accurate dryer simulation.

1. Introduction

A previous work (Giner & Mascheroni, 2001) has shown that the short time simplified analytical diffusion equation developed by Becker (1959) can be applied to thin-layer drying of wheat without losing the accuracy of the standard infinite series solution for spheres, but with a manifest gain in computing speed. This last feature is an advantage in its own right apart from being convenient for interactive drying simulation aimed at design purposes as well as for developing model-based algorithms for drying control (Nellist & Bruce, 1995).

While the computing speed advantage of using Becker's equation in wheat-drying simulation programs is comparable to that obtained with simple empirical and semi-empirical equations as those shown by Jayas *et al.* (1991), the diffusive equation is always as or more accurate and retains physical meaning. Therefore, it is preferable in various respects, among them the possibility of using the effective diffusion coefficients D fitted with Becker's equation (corrected or not by appropriate factors) in more rigorous finite differences (Bruce, 1985) and finite element solutions (Haghighi & Segerlind, 1988; Irudayaraj *et al.*, 1992; Kang & Delwiche, 1999).

The short time expression was used to determine diffusion coefficients in the original Becker's (1959) work, where wheat was dehydrated in a vacuum oven to study the internal moisture migration (the author used a surface moisture independent of drying conditions) and in the opposite situation of liquid water adsorption by wheat (Becker, 1960). Fan *et al.* (1961) and more recently Glenn and Johnston (1994) have also used the short time equation in wheat to fit D in adsorption tests. Concerning forced air drying, Giner and Calvelo (1987) have fitted the short time equation to thin-layer drying wheat curves measured under fluidization conditions at different

Notation

a_v	kernel-specific surface area m^2 m^{-3}
a_{ve}	kernel-specific surface area of the equivalent sphere at a generic moisture content, m^2 m^{-3}
a_{ve0}	value of a_{ve} at the initial moisture content m^2 m^{-3}
d_e	diameter of the equivalent sphere, m
D	effective diffusion coefficient of water in wheat, m^2 s^{-1}
D_∞	Arrhenius pre-exponential factor, m^2 s^{-1}
$D_{\infty 1}$	intercept of the linear correlation of D_∞ with the initial moisture content, m^2 s^{-1}
$D_{\infty 2}$	slope of the linear correlation of D_∞ with the initial moisture content, m^2 kg [dry grain] s^{-1} kg^{-1} [water]
E_a	activation energy for the drying of wheat, J mol^{-1}
G_a	air mass flow rate referred to the tray cross section, kg m^{-2} s^{-1}
h_a	air absolute humidity, kg [vapour] kg^{-1} [dry air]
h_{ra}	relative humidity of drying air, decimal
k_{Page}	kinetic coefficient, s^{-1}
N	exponent
R_g	gas constant, J mol^{-1} K^{-1}
r^2	coefficient of determination
s_y	standard deviation of the estimate (same units as the estimate)
t	time, s
t_f	duration of a drying run, s
T_a	drying air temperature, °C
$T_{a\,abs}$	absolute temperature of drying air, K
W	mean grain moisture content at time t, decimal, d.b.
W_e	equilibrium moisture content of grains, decimal, d.b.
W_0	initial moisture content of grains for the drying experiments, decimal, d.b.
W_{ad}	moisture ratio, defined as $(W - W_e)/(W_0 - W_e)$
W_{adf}	moisture ratio of grains after a drying time t_f

temperatures and one initial moisture content level and then used it as component of a fluidized bed model. A modified version of the program was employed to test the means of improving fluidized bed drying efficiency (Giner & De Michelis, 1988). The original Becker's (1959) parameters were used in fixed-bed drying simulations by Spencer (1969) who then proposed a variable surface grain moisture (Spencer, 1972). Becker's diffusion coefficient correlation combined with a sorption equilibrium equation was used in cross-flow drying simulation by Giner *et al.* (1996). Becker and Sallans (1955) and Sun and Woods (1994) have measured thin-layer drying of wheat in forced air current at a single initial moisture content and used the standard infinite series solution to fit diffusion coefficients at moderate and low temperatures.

The previous literature indicates, on the one hand, that limited thin-layer data of wheat measured in forced convection conditions was used for the purpose of determining kinetic parameters and that there are few drying curves measured where the effect of temperature was isolated from that of initial moisture content (Zogzas & Maroulis, 1996; Zogzas *et al.*, 1994, 1996). On the other hand, the accurate and fast Becker's expression was seldom used to fit diffusion coefficients in drying situations, and, as far as the authors are aware, not to evaluate the effect of initial moisture content. This variable becomes important to test since, according to Maroulis *et al.* (1995), the diffusion coefficient of water in foods may depend on moisture content (m.c.) at low moisture values, where grain drying takes place. Therefore, the purpose of this work is to develop an improved version of the short time Becker's equation mainly for further use in drying simulation programs. To this end, the following specific objectives were set: (1) to contribute experimental data of thin-layer drying of hard wheat measured at several temperatures and starting from different initial moisture contents; (2) to estimate water diffusion coefficients by fitting the short time equation to the data; and (3) to verify the extent of the temperature effect and to devise a practical means of dealing with the possible effects of water concentration on the diffusion equation in an analytical solution while reducing the already small effect of shrinkage.

2. Theoretical considerations

The previous work (Giner & Mascheroni, 2001) has reviewed the development of the analytical short time solution of the diffusion equation for studying wheat-drying kinetics:

$$W_{ad} = \frac{W - W_e}{W_0 - W_e} = 1 - \frac{2}{\sqrt{\pi}} a_v \sqrt{Dt} + 0{\cdot}331\, a_v^2\, Dt \quad (1)$$

where: W is the mean grain moisture content, dec., d.b., while W_0 and W_e are the initial and equilibrium values; and a_v is the kernel-specific surface area. This equation stands for isothermal drying without considering variations of grain volume or changes in the effective diffusion coefficient D with moisture content *during drying*. Therefore, if D depends solely on temperature, Eqn (1) predicts that W_{ad}, the moisture ratio, is a function of time t only. This means that drying runs carried out at the same temperature but starting from different initial moisture contents W_0 are expected to have coincident curves of the moisture ratio. The experimental work that follows here is mainly aimed at plotting the observed curves of W_{ad} *versus* t at the same temperature so as to see whether they coincide or not.

3. Materials and methods

3.1. *Material and moisture content determination*

Registered seed grade hard wheat (*Triticum aestivum*) cv PROINTA- Isla Verde, grown in Pergamino, Provincia de Buenos Aires, Argentina, was used. The wheat was harvested in the 96/97 season and was naturally dried in the field and received free from broken grains and foreign material. Grain moisture content at reception was 12·53% w.b. (0·1433 dec., d.b.). Moistures were measured by the ASAE (1982) whole grain method (19 h at 130°C in forced air oven) using triplicate samples. Hereafter, all grain moistures are expressed in decimal dry basis, except when otherwise stated.

3.2. *Preparation of wheat for the thin-layer drying runs*

The material was divided into four 300 g samples, which were moistened in a rotating drum (Bruce & Giner, 1993; Giner & Denisienia, 1996) adding the necessary amount of water to obtain several initial moisture content values W_0, in a range wider than that usually encountered in wheat drying. Moistened samples were placed in sealed plastic containers and kept for at least 72 h in a cold store at 10°C to allow moisture to distribute inside the kernels while preventing any considerable microbial growth. The values for W_0 obtained were 0·1891, 0·2133, 0·2396 and 0·2694. The containers were transferred from the cold store to the laboratory as needed for the drying experiments, always 1 day before being dried to allow them to equilibrate with room temperature.

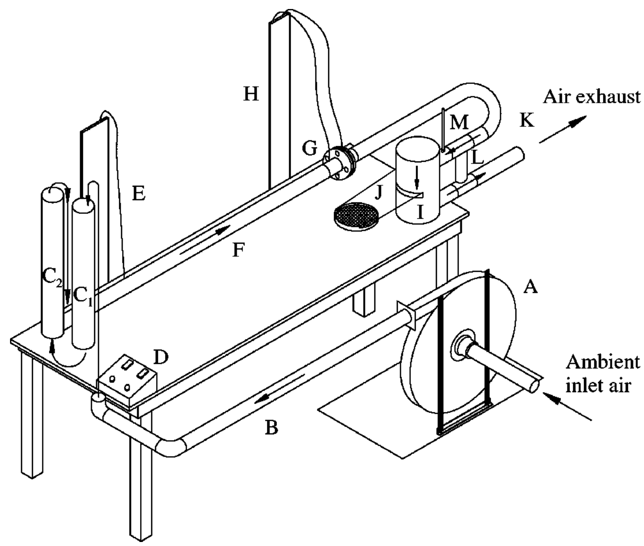

Fig. 1. Schematic of thin-layer drying equipment used in this work; (A), centrifugal fan; (B), air towards the heater; (C_1 and C_2), electrical resistance heaters, 0·6 kW each; (D), power regulation panel; (E), manometric pressure meter; (F), 0·043 m internal diameter duct for heated air; (G), orifice plate; (H), differential pressure meter, (I), drying chamber;(J), 0·175 m mesh-bottomed tray; (K), air exhaust;(L), air bypass; (M), glass thermometer

3.3. *Thin-layer drying equipment*

The equipment used (*Fig. 1*) consisted of a 1·5 kW, 2800 min^{-1} centrifugal fan (A) (maximum flow rate 0·1 m^3 s^{-1}) which drew ambient air from the laboratory and sent it to a tube (B), towards two insulated cylinders each with a 0·6 kW electric resistance (C_1 and C_2) whose power could be regulated (D) to attain the target drying temperature. The air thus heated was passed towards a duct (F) (internal diameter: 0·043 m), and its positive pressure determined using a vertical U tube (E) filled with water, with one end open to the atmosphere. The air was then passed through an orifice meter (G), connected to a differential vertical U tube again filled with water (H) and then directed towards the drying chamber (I) where the air was passed downwards through a 0·175 m internal diameter mesh-bottomed tray (J) that supported the thin layer of grains. Downstream the tray, the air was exhausted (K) sufficiently far from the fan inlet. The system had an air bypass (L) for future 'air-off' weighings *in situ* of the tray (Bruce & Sykes, 1983; Bruce, 1985, 1992) but the automated system was not implemented yet. The drying air temperature at the chamber inlet was measured using a precision glass thermometer (M), accurate to ± 0·5°C.

3.4. *Experimental procedure*

3.4.1. *Weighings and calculation of moistures at different times*

As indicated in Section 3.3, the drying chamber contains a mesh-bottomed tray to support the grains. The tray is removed sideways during the periodic weighings and rapidly replaced. Air leakage from the chamber was prevented during normal drying operation by the appropriate use of weather stripping around the removal/replacement aperture. The removable tray had a 1 cm high wall all around its perimeter to avoid wheat being spilled during handling for weighing, a procedure lasting about 15–20 s. Except the initial value, determined by the oven method, all other moistures of a drying curve were determined using the weighing data, assuming constant dry matter in the tray.

Including the initial weight, each experimental run involved 10 weighings, since more data were considered unnecessary for defining the drying curve besides contributing to disturb the normal evolution of the system. Weighings were done in a precision balance OHAUS Model GT410 (OHAUS Corporation, Florham Park, NJ 07932-0900, readability: 0·001 g). The initial sample weights were around 85 g, and the total weight losses ranged from 6 to 10 g over the 16 drying experiments, so the weighing error was negligible. To check for possible grain spillage or other problems in the drying data, the moisture content of the final grain sample was also determined by the oven method. In effect, dry matter losses were detected in three out of the 16 runs, and these were carried out again and verified further. After this, the mean final moisture difference of values reached by weighing and oven was 0·0023 dec., d.b., which is within the accepted error for oven moisture determination (ASAE, 1982).

3.4.2. *Advance approximate planning of experimental drying times and weighings distribution*

Bruce (1985) has indicated that, for parameter-fitting purposes, different thin-layer experiments should have the same final moisture ratio W_{adf}. To have an estimate of the drying time t_f required to reach W_{adf}, for each drying temperature, preliminary drying experiments were done where commercial wheat provided by a local miller was rewetted to an initial moisture content W_0 of 0·2646 and dried at 40, 50 and 60°C. The phenomenological Page equation (Jayas *et al*., 1991), whose expression is $W_{ad} = \exp(-k_{Page}\, t^N)$ was used to fit the data. The value of the exponent N was 0·60, while the kinetic coefficient in s^{-1} was expressed as $k_{Page} = 34{\cdot}6 \exp(-2820/(T_a + 273{\cdot}16))$. Then, by choosing a value for W_{adf} of 0·3, estimates of $t_f = (-\ln W_{adf}/k_{Page})^{1/N}$ were obtained which were rounded to 240, 125, 80 and 60 min for drying air temperatures T_a of 35, 50, 60 and 70°C, respectively. These values are typical in the heated-air drying of wheat (Nellist & Bruce, 1995).

3.4.3. *Maintenance of the drying air temperature*

In each experiment, drying air temperatures at the chamber inlet were kept constant by manual regulation of the power of each electrical resistance (see *Fig. 1*), by visually monitoring the thermometer at the drying chamber inlet and by further fine adjustment of electrical power when necessary. By this method, the drying air temperature was not observed to depart by more than $\pm$ 0·5°C of the target value in all drying experiments.

3.4.4. *Airflow determination*

The flow of air towards the drying chamber was determined from the pressure drop across a 40 mm concentric-type orifice meter for gases (maximum flow of 110 kg h^{-1}).

3.4.5. *Determination of ambient air conditions*

In each experiment, a psychrometer was used to take several readings of dry bulb and wet bulb temperatures along each run, and the averages (variations were very small) employed to calculate the air absolute humidity h_a through 'VAPAIR', a previously developed computer program of the psychrometric chart (Giner, 1999).

3.4.6. *Air mass flow rate chosen for the thin-layer drying experiments*

Based on the analysis carried out in the previous work (Giner & Mascheroni, 2001), values for the air mass flow rate G_a of around 0·3 kg m^{-2} s^{-1} were chosen because they allow a predominantly external control for heat transfer while ensuring strict internal control for mass transfer. Besides, this flow rate is a practical value used

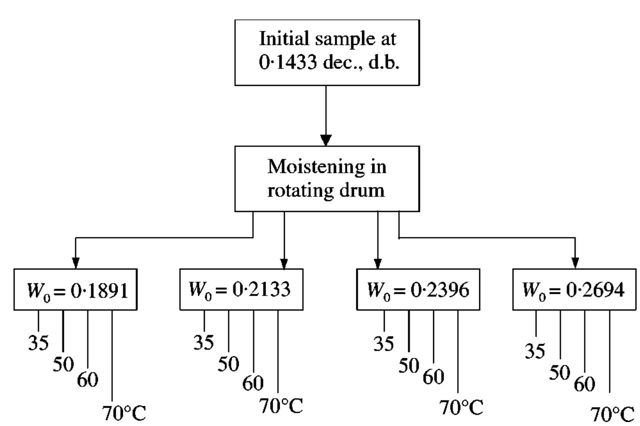

Fig. 2. Diagram showing samples prepared with different initial moistures W_0 and dried at four temperatures

Table 1

Operating conditions for thin-layer drying experiments in hard wheat; the equilibrium moisture content $W_e = f(T_a, h_{ra})$ was based on the Modified Henderson isotherm equation for hard wheat (Giner & Mascheroni, 2001); the air mass flow rate G_a was referred to as the tray cross section; m.c., moisture content

Initial grain, m.c. (W_0), dec., d.b.	*Drying air temperature (T_a), °C*	*Ambient dry bulb temperature, °C*	*Ambient wet bulb temperature, °C*	*Absolute humidity (h_a), kg kg^{-1}*	*Relative humidity of drying air (h_{ra}), decimal*	*Equilibrium moisture content (W_e), dec., d.b.*	*Air mass flow rate (G_a), kg m^{-2} s^{-1}*
0·2694	35	24·5	19·6	0·012	0·35	0·103	0·30
	50	20·0	18·6	0·013	0·17	0·066	0·33
	60	19·0	18·1	0·013	0·10	0·050	0·35
	70	19·1	18·0	0·012	0·06	0·039	0·37
0·2396	35	19·0	17·9	0·012	0·35	0·103	0·31
	50	20·3	18·7	0·013	0·17	0·066	0·32
	60	20·8	18·7	0·013	0·10	0·050	0·35
	70	25·7	20·0	0·012	0·06	0·039	0·35
0·2133	35	19·4	16·3	0·010	0·29	0·094	0·30
	50	21·8	17·8	0·011	0·14	0·062	0·33
	60	22·0	18·0	0·011	0·09	0·048	0·35
	70	26·7	20·5	0·013	0·07	0·040	0·35
0·1891	35	22·0	18·7	0·012	0·34	0·102	0·31
	50	23·5	20·2	0·014	0·17	0·068	0·32
	60	24·5	21·0	0·014	0·11	0·054	0·34
	70	24·8	19·8	0·012	0·06	0·039	0·35

in dryers and still high enough to ensure constant air conditions in a thin layer one-grain deep (Woods & Favier, 1993).

3.5. *Experimental programme*

Each of the four samples prepared to a specific moisture content was subdivided into another four for drying tests at 35, 50, 60 and 70°C. The experimental programme is summarized in *Fig. 2*.

4. Results and discussion

Operating conditions for the 16 drying experiments are listed in Table 1.

4.1. *Effect of air temperature*

Experimental results of the mean grain moisture W in the drying curves were expressed in dimensionless form as the moisture ratio $W_{ad} = (W - W_e)/(W_0 - W_e)$ (see data of Table 1). The influence of air temperature was assessed by grouping the data by level of initial moisture content. Results are shown in *Fig. 3*.

In each graph of *Fig. 3*, the data exhibit a well-defined falling–drying rate since the slope reduces continuously with time. On the other hand, while it is known that the increase of temperature strongly speeds up the drying, it may be interesting to give an example based on experimental data since, as inferred from a review by Jayas *et al.* (1991), there is no general agreement on how strong the effect is. For instance, at a value for W_0 of 0·2694 [*Fig. 3*(*a*)] the times required to reduce the value of W_{ad} from 1 to $\approx$ 0·35 were about 180, 105, 80 and 50 min for air temperatures of 35, 50, 60 and 70°C, respectively, showing how temperature reduces drying time. These were comparable with the estimates of the Page equation mentioned above. The temperature effect was similar in the graphs at initial moisture contents of 0·2396, 0·2133 and 0·1891 [*Fig. 3*(*b*), 3(*c*), 3(*d*), respectively].

Besides, there appear to be some differences among the final moisture ratios W_{adf} of *Fig. 3* when compared at the same drying time: these tend to be lower for higher initial moisture contents. The values of W_{adf} are between 0·25 and 0·35 for W_0 of 0·2694 [*Fig. 3*(*a*)] while they fluctuate around 0·35–0·45 for W_0 of 0·1891 [*Fig. 3*(*d*)]. Therefore, the mean drying rate seems to be higher, even in dimensionless terms, when drying starts from higher values of W_0. To verify the possible effect of W_0, the data are regrouped by temperature level.

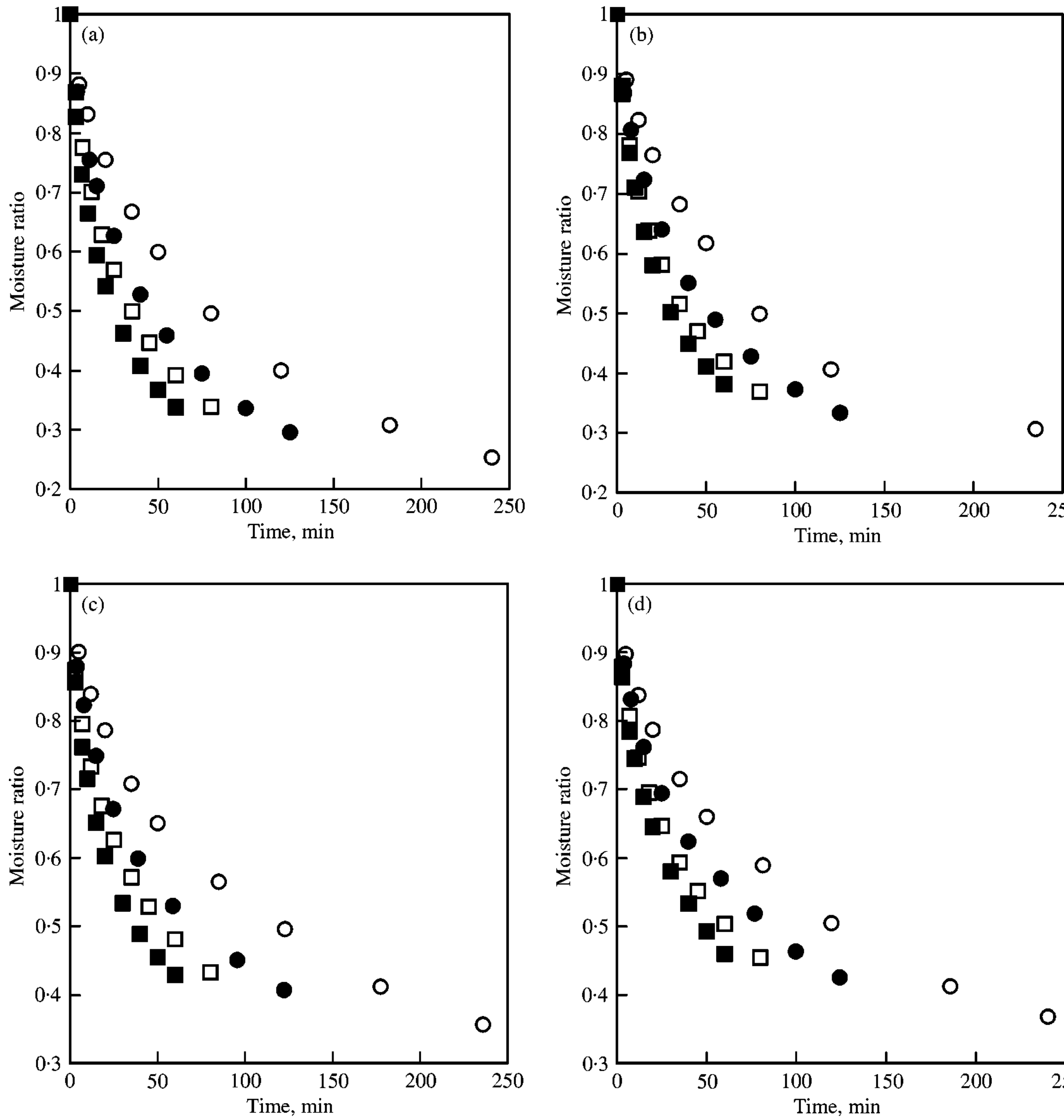

Fig. 3. Experimental moisture ratios of grains as a function of time for air temperatures of 35 (○); 50 (●); 60 (□) and 70°C (■) with reference to four initial moisture contents (dec., d.b.): (a) 0·2694; (b) 0·2396; (c) 0·2133 and (d) 0·1891

4.2. *Effect of the initial moisture content*

Unlike the effect of temperature on drying rate (and on the diffusion coefficient) (Sokhansanj & Cenkowski, 1988; Jayas *et al.*, 1991; Nellist & Bruce, 1995), the effect of moisture content on the drying curve is not sufficiently studied yet and the results collected on grains are scarce and not conclusive (Zogzas *et al.*, 1994, 1996; Zogzas & Maroulis, 1996). To this end, the data measured here were grouped by temperature to observe any possible direct influence of W_0. It must be said that although the increase of W_0 enlarges the mass transfer driving force ($W_0 - W_e$) causing a speeding-up effect on the dimensional drying rate ($-\,dW/dt$), this is compensated when moisture is expressed in dimensionless form as the moisture ratio.

At 35°C [*Fig. 4*(*a*)], however, compensation is not complete for the W_{ad} *versus* t curves depart as drying progresses, becoming 'faster' (falling more rapidly even though moisture content is expressed as the moisture ratio) when starting from higher values of W_0. Graphs grouped at 50°C [*Fig. 4*(*b*)], 60°C [*Fig. 4*(*c*)] and 70°C [*Fig. 4*(*d*)] also corroborate this behaviour. Therefore, apart from the effect of W_0 on the driving force, a consistent additional effect of water content is observed which should be ascribed to the internal water transport characteristics of the grain.

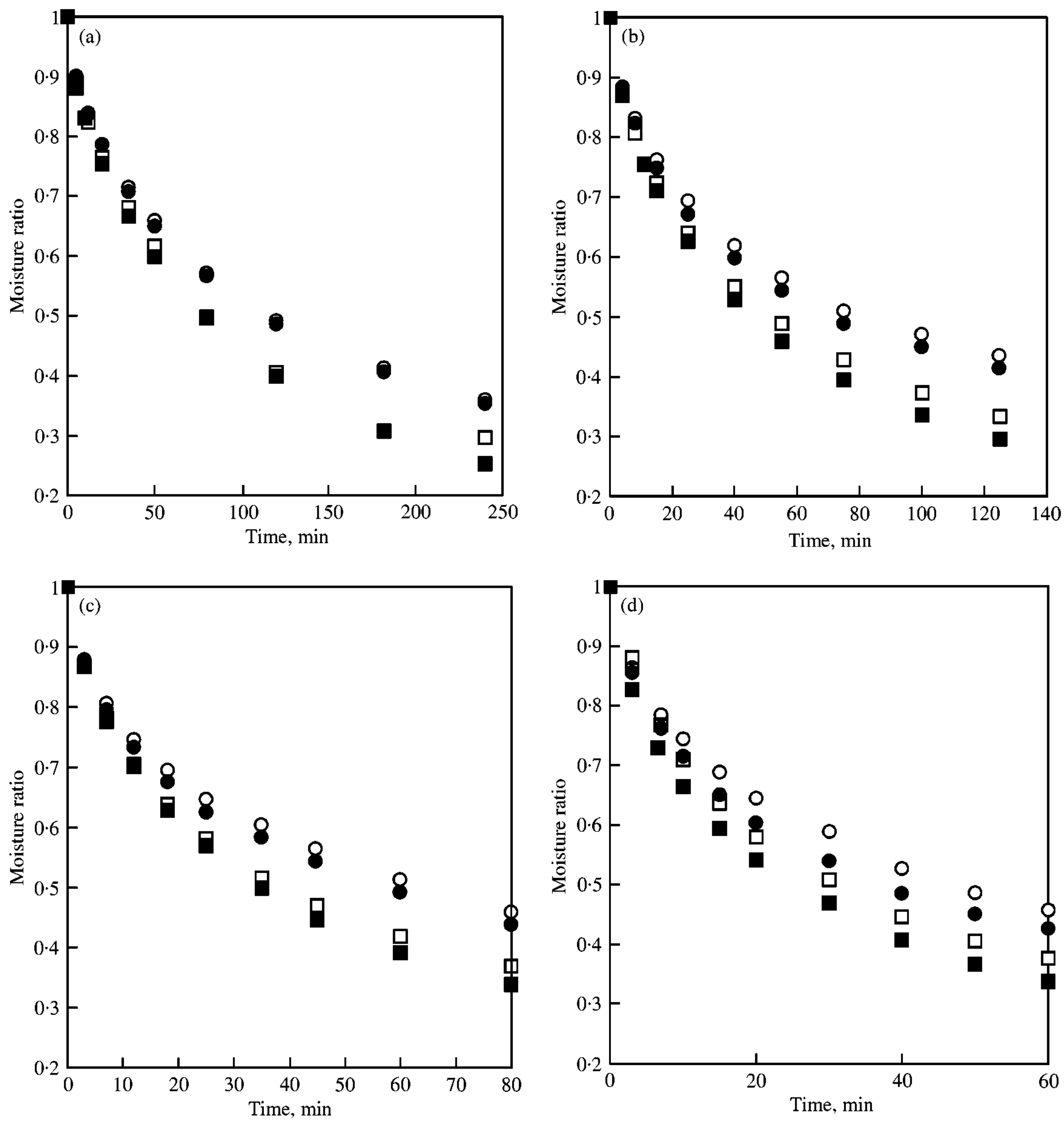

Fig. 4. Experimental moisture ratios of grains as a function of time for initial moisture contents of 0·2694 dec., d.b. (■); 0·2396 (□); 0·2133 (●) and 0·1891 (○); data were grouped by air temperature: (a) 35, (b) 50, (c) 60 and (d) 70°C

As indicated above in Section 2, the analytical solutions predict coincident curves of moisture ratio *versus* time when drying at the same temperature regardless of the initial moisture content, so the results observed in *Fig. 4* should be atributted to some increase of the diffusion coefficient with moisture content. The rest of the paper shows the fitting of the short time equation to every experimental drying run, and the development of a correlation of the diffusion coefficient that allows for the effect of both temperature and initial moisture content.

4.3. *Fitting the short time equation to thin-layer drying data of wheat*

To apply Eqn (1) to the W_{ad} *versus* t data of the 16 thin- layer drying experiments, these were grouped by initial moisture content level and the fittings carried out run by run at the different temperatures. It must be indicated that no data were taken for $W_{ad} < 0{\cdot}2$, so all moistures can be interpreted with the short time equation.

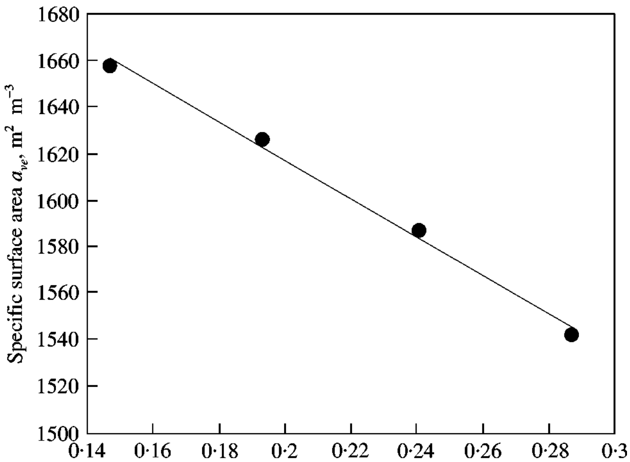

Fig. 5. Equivalent spherical surface area of grain (●), as a function of moisture content; the linear regression (line) had a coefficient of determination r^2 of 0·994

4.3.1. *Use of specific grain surface dependent on initial moisture*

Grain-specific areas (particle surface area/particle volume) were those of the sphere having the same volume of the kernel. The pycnometric wheat volume was measured on the same variety as a function of m.c. in a previous work (Giner & Denisienia, 1996), and the data were used to calculate the equivalent spherical diameter d_e and then the equivalent specific area a_{ve} as $6/d_e$. A linear correlation between a_{ve} and moisture content ($a_{ve} = 1781{\cdot}2 - 820{\cdot}1W$) was obtained with a coefficient of determination r^2 of 0·994. Values and correlation of a_{ve} are shown in *Fig. 5*.

The graph shows that a_{ve} varies some 6% over a moisture range wider than that of the usual wheat drying. To reduce the already small effect of using only one value of a_{ve} in the analytical solution *during drying*, the specific surface areas were calculated at the initial moisture content in each run W_0 and were noted as a_{ve0}.

4.3.2. *Preliminary indirect fitting of the short time equation to experimental data*

For each initial moisture content, the indirect fitting consists of two steps: (1) to fit the short times equation [Eqn (1)] to drying curves at the several temperatures, to obtain the corresponding diffusion coefficients *D*; and (2) to correlate the values for *D* with temperature (Sun & Woods, 1994). In this study, there are four levels of W_0 so step (2) is done 4 times. Finally, the relationship between the parameters of step (2) and W_0 is investigated. Table 2 shows the results of the fittings of all drying curves; the table also contains the final moisture content reached in each run as well as statistical estimates of goodness of fit (Wilkinson, 1990).

At each level of W_0, Table 2 shows that the short time equation fits very closely the experimental drying data in each run (step 1), with low standard deviations of the

Table 2

Effective diffusion coefficients *D* of water in wheat grains fitted with the short time equation using a kernel-specific surface area dependent on the initial moisture content (m.c.); statistical parameters of goodness of fit as the coefficient of determination r^2 and the standard deviation of the estimate s_y, are included

Initial grain, m.c. (W_0), dec., d.b.	*Kernel-specific surface area (a_{ve0}), m^2 m^{-3}*	*Drying air temperature (T_a), °C*	*Final grain moisture content (W_f), dec., d.b.*	*Effective diffusion coefficient $D \times 10^{-11}$, m^2 s^{-1}*	r^2	s_y
0·2694	1560	35	0·1450	2·278 (± 0·083)	0·996	0·017
		50	0·1261	3·861 (± 0·111)	0·996	0·015
		60	0·1246	5·083 (± 0·139)	0·996	0·014
		70	0·1170	7·111 (± 0·139)	0·997	0·012
0·2396	1585	35	0·1436	1·986 (± 0·096)	0·990	0·023
		50	0·1238	3·250 (± 0·083)	0·997	0·013
		60	0·1202	4·444 (± 0·111)	0·997	0·013
		70	0·1155	5·458 (± 0·212)	0·990	0·048
0·2133	1606	35	0·1360	1·472 (± 0·028)	0·997	0·011
		50	0·1222	2·417 (± 0·055)	0·997	0·011
		60	0·1193	3·333 (± 0·083)	0·997	0·011
		70	0·1114	4·611 (± 0·014)	0·994	0·014
0·1891	1626	35	0·1334	1·389 (± 0·028)	0·998	0·010
		50	0·1189	2·111 (± 0·055)	0·997	0·010
		60	0·1152	2·889 (± 0·055)	0·998	0·008
		70	0·1080	3·667 (± 0·055)	0·999	0·006

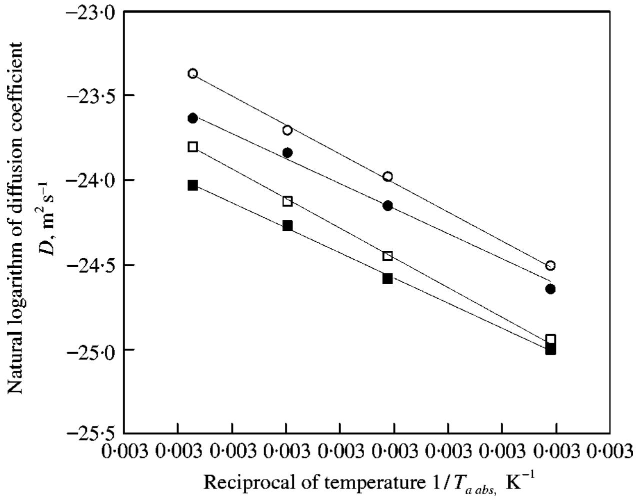

Fig. 6. Semi-logarithmic graph of the diffusion coefficients obtained with the short time equation versus the reciprocal of the absolute air drying temperature $T_{a\,abs}$, grouped by initial moisture contents (W_0) levels: (○), 0·2694, dec., d.b.; (●), 0·2396; (□), 0·2133; (■), 0·1891. Corresponding solid lines show the Arrhenius predictions

estimate s_y and high coefficients of determination r^2, the values of D being obtained with low standard deviations. The values of s_y, a measure of the average deviations between predicted and experimental moisture ratios, were calculated following the definition given in a previous work (Giner & Denisienia, 1996). The results of Table 2 confirm the capability of the short time equation as a kinetic expression and by extension of the liquid diffusion theory for wheat drying when applied to these ranges of temperature and moisture content.

4.3.3. *Preliminary correlation of diffusion coefficients with temperature*

The values for D in Table 2 were plotted *versus* temperature in the Arrhenius form, ln D *versus* $1/T_{a\,abs}$, $T_{a\,abs}$ being the air temperature in K, and a definite linear behaviour was observed (*Fig. 6*) in each initial moisture content level. To fit the Arrhenius expression, both exponential and logarithmic forms were tested:

$$D = D_\infty \exp\left(-\frac{E_a}{R_g T_{a\,abs}}\right) \qquad (2)$$

$$\ln D = \ln D_\infty - \frac{E_a}{R_g T_{a\,abs}} \qquad (3)$$

where D_∞ is the pre-exponential factor and E_a the activation energy. The exponential form was chosen because it was slightly more accurate (though D_∞ and E_a were very close), and the values obtained are listed in Table 3. Nellist and Bruce (1995) have indicated that small changes in the activation energy motivate large changes in the pre-exponential factor, casting doubts on the significance of these parameters when fitted in a narrow absolute temperature range. Table 3 shows that, in effect, this is the case with D_∞ but the fact that E_a was estimated at four levels of W_0 without any large variation around an average of some 27·2 kJ mol^{-1} is a sign that a representative 'activation energy band' was found for wheat. At each level of W_0, the r^2 values indicate that the Arrhenius relationship fits very well the dependency of the diffusion coefficients obtained in Table 2 with temperature. The Arrhenius prediction is also included for each W_0 in *Fig. 6.*

4.3.4. *Effect of initial moisture content on the diffusion coefficient*

After finding a very good Arrhenius fit for the D *versus* T_a data at each initial moisture content level, the next step is to analyse the dependency of the Arrhenius parameters, D_∞ and E_a with initial moisture content. Table 3 shows that D_∞ and, to a much lesser extent, E_a vary irregularly with W_0, so further modelling of these parameters as functions of the initial moisture content appears difficult at first glance. Nonetheless, the four Arrhenius relationships plotted together in *Fig. 6* show a definite vertical shift with no crossovers for increasing values of W_0. Then, despite the irregularities mentioned above, the diffusion coefficient, on the whole, increases with W_0. Besides, as *Fig. 6* also shows that the slope, proportional to E_a, does not present any definite tendency with W_0, the Arrhenius fit was reconsidered by using first an arithmetic average of the activation energy

Table 3
Preliminary values of the Arrhenius pre-exponential factor D_∞ and energy of activation E_a for heated air wheat drying; goodness of fit statistics are also listed; m.c., moisture content

Initial grain m.c. (W_0) dec., d.b.	*Pre-exponential factor $D_\infty \times 10^{-7}$, m^2 s^{-1}*	*Activation energy (E_0), kg mol^{-1}*	*Coefficient of determination (r^2)*
0·2694	16·01 ($\pm$ 6·362)	28·634 ($\pm$ 1107·9)	0·998
0·2396	3·197 ($\pm$ 1·837)	24·720 ($\pm$ 1597·0)	0·993
0·2133	12·88 ($\pm$ 3·047)	29·242 ($\pm$ 659·74)	0·999
0·1891	2·115 ($\pm$ 0·610)	24·712 ($\pm$ 801·8)	0·998

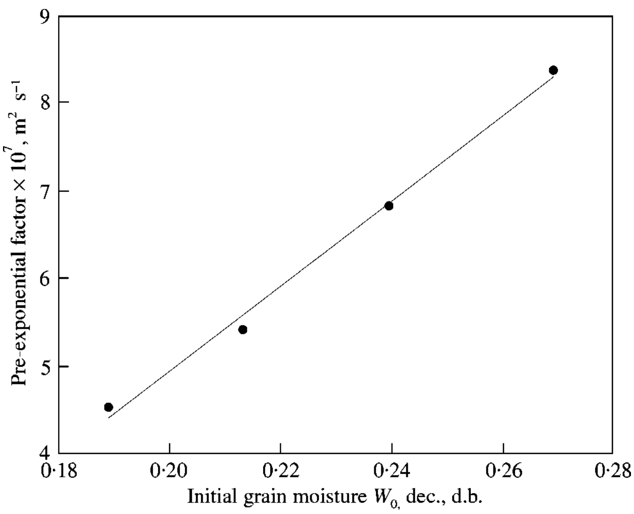

Fig. 7. Trend of the Arrhenius pre-exponential factor (D_∞) with the initial moisture content; symbols (●) are values fitted for an average activation energy of 26·826 kg mol^{-1}. The linear behaviour (solid line) is clear

(26·826 kJ mol^{-1}) to explore the behaviour of D_∞ with W_0. *Figure 7* shows that D_∞ now shows a very good increasing linear correlation with W_0. The preliminary conclusions of the indirect fitting are: (i) that the short times diffusion equation [Eqn (1)] fits very well every thin-layer drying curve; (ii) that, at each level of W_0, the values for D fitted in (i) correlate very well with air temperature showing an Arrhenius behaviour; and (iii) that there is a vertical upwards shift of the Arrhenius correlation for increasing initial moisture content, this shift being better represented using an average constant slope (*i.e.* an average activation energy) and a linear increase of the pre-exponential factor with initial moisture content. The functional correlation of D with initial moisture allows the short time analytical solution—which assumes that D does not vary with moisture content *during drying*—to be used with a constant value of D for each W_0 yet with the possibility of estimating a different value of D for different W_0.

The next item is to combine the results of the three steps of indirect fitting to directly apply the short time diffusive equation to the whole thin-layer drying data pool, collected as described in Section 3, so as to find the final values of the parameters.

4.3.5. *Direct fitting of the short time diffusive model to wheat thin-layer drying curves*

The direct fitting was carried out by applying Eqn (1) now rewritten in dimensional form as

$$W = W_e + (W_0 - W_e)$$
$$1 - \frac{2}{\sqrt{\pi}} a_{ve0} \sqrt{Dt} + 0{\cdot}331\, a_{ve0}^2\, Dt \quad (4)$$

together with

$$D = [D_{\infty 1} + D_{\infty 2}(W_0 - 0{\cdot}1891)] \exp\left(-\frac{E_a}{R_g T_{a\,abs}}\right) \quad (5)$$

(the correlation should not be used below the lowest experimental W_0 tested here, i.e. 0·1891) and

$$a_{ve0} = 1781{\cdot}2 - 820{\cdot}1 W_0 \quad (6)$$

to fit the complete experimental data set of t, W, W_0 and T_a directly. In Eqn (5), $D_{\infty 1}$ is the intercept and $D_{\infty 2}$ the slope of the linear correlation of D_∞ with the initial moisture content, and R_g is the gas constant. A total of 159 t–W pairs were used to estimate three fitting parameters: $D_{\infty 1}$, $D_{\infty 2}$ and E_a, so the degrees of freedom result 156. The final results of the fitting were: $D_{\infty 1}$, 5·046 ($\pm$ 1·088) × 10^{-7} m^2 s^{-1}; $D_{\infty 2}$, 54·44 ($\pm$ 10·58) × 10^{-7} m^2 s^{-1} while E_a, 27·184 ($\pm$ 523·8) kJ mol^{-1}. The overall coefficient of determination r^2 was of 0·996 with a standard deviation of the estimate s_y of 0·0025 dec., d.b., *i.e.* in the order of errors in oven moisture determination, so the predictions are very good for the whole set of experimental data collected here.

4.4. *Comparison of observed thin-layer data with predicted curves*

Although the values for r^2 and s_y described above were both very good for the fitting of the combined model [Eqns (4), (5) and (6)] to the whole thin-layer data pool, and they may be enough to assure the good behaviour of the simplified diffusive equation for short times, some experimental grain m.c. data were plotted in *Fig. 8* as a function of time together with the corresponding predicted curves. To this end, three drying experiments were selected which differ from each other both in drying temperature and initial moisture content. Predicted curves are observed to reproduce very well the experimental drying kinetics of wheat with respect to the influence of time, drying air temperature, air humidity, and grain initial moisture content, showing the suitability of the short time diffusive expression.

4.5. *Comparison of the diffusion coefficient with previous research*

From reviews by Zogzas *et al.* (1994, 1996) and Zogzas and Maroulis (1996), it was observed that diffusion coefficients can vary noticeably from one author to another, even for the same food. For that reason, the values of D obtained here were calculated as a function of temperature for W_0 of 0·19 and 0·25, and compared with those

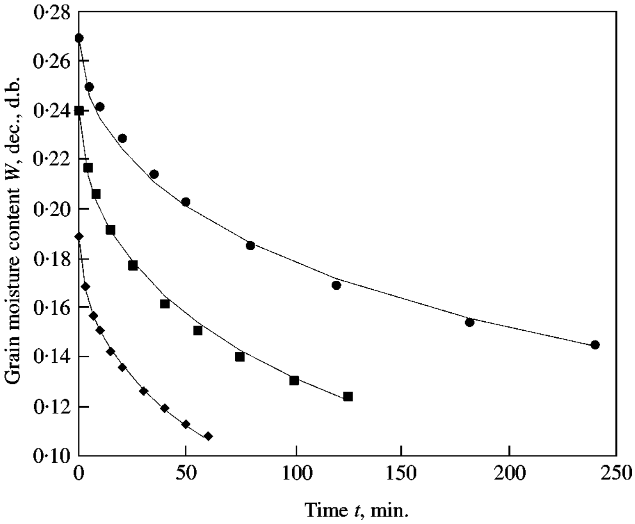

Fig. 8. Moisture content of wheat as a function of time for three conditions that differ both in initial moisture content and temperature: 0·2694 dec, d.b. and 35°C (●); 0·2396 and 50°C (■); 0·1891 and 70°C (◆). Solid lines show the corresponding predictions of the combined kinetic model

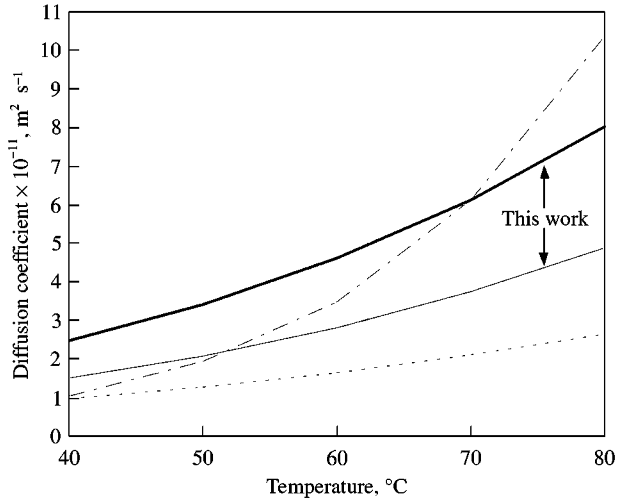

Fig. 9. Calculated water diffusion coefficients in wheat plotted versus temperature. Predictions of the correlation obtained in this work are shown for initial moisture contents of 0·25 dec., d.b. (——) and 0·19 (——). The graph also includes correlations by Becker (1959) (– · –) and Sun and Woods (1994) (· · · ·)

presented by Sun and Woods (1994) and Becker (1959) for wheat, which do not include the effect of initial moisture on D.

The literature correlations for D in $m^2 s^{-1}$ for studies of wheat by Becker (1959) and Sun and Woods (1994) are, respectively:

$$D = 76{\cdot}8 \times 10^{-4} \exp\left(-\frac{51\,081}{R_g T_{a\,abs}}\right) \tag{7}$$

$$D = 7{\cdot}51 \times 10^{-8} \exp\left(-\frac{23\,447}{R_g T_{a\,abs}}\right) \tag{8}$$

The predictions of Eqns (7) and (8) are plotted in *Fig. 9* together with those of Eqn (5).

The diffusion coefficients obtained here experience a twofold increase when changing the initial moisture content from W_0 of 0·19 to 0·25. In turn, they are 2–3 times as high as those obtained by Sun and Woods (1994), with a comparable, but stronger temperature dependency. Becker's (1959) values of D show the effect of his high activation energy ($E_a = 51{\cdot}081$ kJ mol^{-1}). Values obtained here are higher than Becker's at low temperatures but not at higher temperatures, where the opposite occurs. As the values for D fitted here are calculated at higher W_0 the crossover with Becker's diffusion coefficients shifts to higher temperatures. Apart from the influence of the initial moisture content, differences among diffusion coefficients can originate from differences in wheat varieties (for instance, the specific surface area assigned to the grain), in the techniques used to collect thin-layer data, and, most possibly, in the equilibrium moisture content W_e used. Values of D are affected by W_e since the latter is used to express the experimental m.c. as a dimensionless variable: the moisture ratio.

The value of E_a obtained in this work (≈ 27 kJ mol^{-1}) is in reasonable agreement with that found by Sun and Woods (1994) ($\approx 23{\cdot}5$ kJ mol^{-1}) and very similar to an apparent activation energy found by Bruce (1985) for barley (≈ 26 kJ mol^{-1}). In Becker's (1959) data, as mentioned above, his value of some 51 kJ mol^{-1} must have been influenced by the use of a surface moisture content of 0·103 dec., d.b., independent of temperature and relative humidity. Hence, as his W_e value was high, the driving force became reduced, so a higher value of D was required for compensation, particularly at high temperatures.

The results obtained here contribute to the area of diffusion coefficients determination since former reviews of grain-drying kinetic parameters (Sokhansanj & Cenkowski, 1988; Jayas *et al*., 1991) or diffusion coefficients in foods (Zogzas *et al*., 1994, 1996; Zogzas & Maroulis, 1996) have not found reliable correlations of the diffusion coefficient with the initial moisture content. Therefore, the version of Becker's equation developed here can be used as a thin-layer equation in fixed bed or continuous dryer simulation programs for wheat.

5. Conclusions

An experimental programme was followed to measure thin-layer drying curves of wheat which allowed the effect of air temperature to be isolated from that of initial moisture content.

The experimental curves of moisture ratio *versus* time, grouped by initial moisture level, showed the expected strong speeding-up effect of temperature on drying rate. When these curves were grouped by temperature, the dimensionless drying curves tended to depart from each other as drying proceeded, falling faster when starting from higher initial moisture contents. This is an evidence of the dependency of the effective diffusion coefficient of water in wheat with moisture content.

The short time diffusive solution fitted the kinetic data very closely and reproduced well the shape of the experimental drying curve. The only fitting parameter is the effective diffusion coefficient. Some account for shrinkage was taken by including a grain-specific surface area variable with the initial moisture content.

At each initial moisture content, the diffusion coefficient followed very well an Arrhenius-type behaviour. Diffusion coefficients increased steadily with the initial moisture content and this was best accounted for by a linear increase of the pre-exponential factor keeping the energy of activation constant (about 27 kJ mol^{-1}).

This short time analytical diffusive equation for small grains is recommended for use in drying simulation programs aimed at design or control, which require accuracy and calculation speed.

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