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Bean moisture diffusivity and drying kinetics: a comparison of the liquid diffusion model when taking into account and neglecting grain shrinkage

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Abstract

The aim of this work was to compare the results of the liquid diffusion model with respect to bean (*Phaseolus vulgaris* L.) drying when taking into account or neglecting grain shrinkage. Bean grains were harvested with a moisture content of 0.92 kg water/kg dry matter and dried at air temperatures of 25-55°C and relative humidities of 20-75%. The volume of each grain, understood as a sphere, was determined several times over the drying process, taking the diameter to be the mean length of the three orthogonal axes. Grain shrinkage was determined by examining the relationships between the volume associated with each moisture content and the initial volume. The results show that the liquid diffusion model describes the drying kinetics of beans satisfactorily, and that grain shrinkage can be ignored. The diffusion coefficient increases with air temperature, with values ranging between 10.8×10^{-10} and 67.0×10^{-10} m^2 s^{-1}. This is described by the Arrhenius equation, with an activation energy of 40.08 kJ mol^{-1}.

Additional key words: diffusion coefficient, grains, mathematical models, moisture, *Phaseolus vulgaris*.

Resumen

Difusión de la humedad y cinética del secado de judías: comparación de la validez del modelo de difusión líquida, incluyendo y excluyendo la pérdida de volumen

El objetivo de este trabajo fue ajustar el modelo de difusión líquida para predecir las pérdidas de volumen de grano en el secado de judía (*Phaseolus vulgaris* L.). Se recolectaron granos de judía con una humedad del 0,92 y se sometieron a un proceso de secado, bajo condiciones controladas, a diferentes temperaturas entre 25 y 55°C y humedades relativas entre 20 y 75%. El volumen de cada grano, considerado como una esfera, fue obtenido utilizando como diámetro el promedio de las tres diagonales principales durante el proceso del secado. Las pérdidas de volumen de grano se determinaron por la relación entre el contenido de agua inicial y final. A partir de los resultados obtenidos, se concluye que el modelo de difusión representa satisfactoriamente la cinética del secado de la judía, y que tiene en cuenta las pérdidas de volumen del grano. El coeficiente de difusión, con valores comprendidos entre $10{,}8 \times 10^{-10}$ y $67{,}0 \times 10^{-10}$ m^2 s^{-1}, aumenta con la temperatura. La variación del coeficiente de difusión con la temperatura puede ser descrita por la expresión de Arrhenius, con una energía de activación de 40,08 kJ mol^{-1}.

Palabras clave adicionales: coeficiente de difusión, grano, humedad, modelos matemáticos, *Phaseolus vulgaris*.

Introduction

Water is the main component of most agricultural products and it has enormous influence on their physical properties. Agricultural products are usually dried to ensure their quality and stability. Diminishing moisture levels lead to a reduction in biological activity and induce chemical and physical changes in grains.

According to Prado *et al.* (2000), the removal of water during the drying of grains leads to a reduction in the tension inside cells, promoting product shrinkage. The majority of the models used to predict the drying

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of agricultural products were developed without taking into account the importance of volumetric contraction during dehydration (Brooker *et al.*, 1992). According to Ramos *et al.* (2005), this should be included in models for a complete description and analysis of the phenomenon to be made.

Drying models based on the liquid diffusion theory have attracted special attention from researchers. The liquid diffusion mechanism is extremely complex due to the diversity of the chemical composition and physical structure of different products. In addition, the use of different experimental methodologies renders comparisons difficult.

According to Brooker *et al.* (1992), Fick's second law is used in liquid diffusion theory to establish moisture diffusion as a function of the concentration gradient. The following equation describes a one dimensional drying situation (in Cartesian coordinates):

$$\frac{\partial U^*}{\partial t} = \frac{\partial}{\partial x}\left(D \cdot \frac{\partial U^*}{\partial x}\right) \quad [1]$$

where U^* is the product moisture content (kg water/kg dry matter), t is the time (in s), x is the distance between two points of reference in the product (in m), and D is the liquid diffusion coefficient (in $m^2\ s^{-1}$).

The variation in moisture content as a function of the drying time in homogeneous matter with a constant diffusion coefficient is represented by the following equation (Brooker *et al.*, 1992):

$$\frac{\partial U^*}{\partial t} = D \cdot \left[\frac{\partial^2 U^*}{\partial r^2} + \frac{c}{r} \cdot \frac{\partial U^*}{\partial r}\right] \quad [2]$$

where r is the radial distance (in m), and c = «0» for flat bodies, «1» for cylindrical bodies and «2» for spherical bodies.

Many solutions to Equation [2] for different geometric shapes have been used to describe the process of drying in agricultural products in boundary conditions (Brooker *et al.*, 1992):

$$U^*(r, 0) = U_i^* \quad [3]$$

$$U^*(r, t) = U_e^* \quad [4]$$

where U_i^* is the initial moisture content (kg water/kg dry matter), and U_e^* is the moisture content equilibrium (kg water/kg dry matter).

Brooker *et al.* (1992) provided the analytical solution to Equation [2] for spherical shapes as follows:

$$MR = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left[-\frac{n^2 \cdot \pi^2 \cdot D \cdot t}{9} \cdot \left(\frac{3}{r}\right)^2\right] \quad [5]$$

where MR is the product moisture ratio (dimensionless), and n the number of terms.

The analytical solution of these equations is based on an infinite series, with the number of terms determining the precision of the results. Afonso-Júnior and Corrêa (1999) evaluated the results of drying the bean (*Phaseolus vulgaris* L.) cultivar Ouro Negro using the diffusion model adjusted for a spherical geometry and by approximating with a series of 8 terms. This was found to be adequate and provided satisfactory estimates of the product drying rate.

During the modelling and simulation of the drying of agricultural products, several authors have satisfactorily correlated the diffusion coefficient with different drying variables (Madamba *et al.*, 1996; Afonso Júnior and Corrêa, 1999; Ozdemir and Devres, 1999; Doymaz, 2005; Mohapatra and Rao, 2005).

The diffusion coefficient generally increases as the air temperature increases (Ramesh, 2003), and can be described by the Arrhenius equation as a function of temperature:

$$D = D_o \cdot \exp\left(\frac{-E_a}{R \cdot T_a}\right) \quad [6]$$

where D_o is the pre-exponential factor (in $m^2\ s^{-1}$), E_a is the activation energy (in kJ mol^{-1}), R the universal gas constant (8,314 J $mol^{-1}\ K^{-1}$), and T_a the absolute temperature (in ºK).

Arrhenius expression coefficients are easily obtained by making the equation linear, applying the following logarithm:

$$\ln D = \ln D_o - \frac{E_a}{R} \cdot \frac{1}{T_a} \quad [7]$$

Due to the importance of the study of the drying of tropical agricultural products, and the lack of theoretical information regarding the phenomena that occur during bean drying, the aim of this work was to compare the results of the liquid diffusion model under different conditions, taking into account or neglecting grain shrinkage.

Material and Methods

The beans used in this work were of the red group, cv. Vermelhinho. These were manually harvested at a moisture content of around 0.92 kg water/kg dry matter. All experiments were performed at the Laboratory of Physical Properties and Quality of Agricultural Products-CENTREINAR, *Universidade Federal de Viçosa*, Viçosa-MG, Brazil.

Product moisture was determined using the greenhouse method (MARA, 1992), leaving the beans at 105 ± 1°C for 24 h until a constant mass was achieved.

Drying experiments were performed in a controlled environment chamber (Aminco) under different conditions of temperature (25, 35, 45 or 55°C) and relative humidity (20 or 75%). The total number of combinations of conditions was 14. Drying proceeded until the product reached its equilibrium moisture content under the conditions set.

Two removable trays —each containing 50 g of beans— with netted bottoms to allow air to pass through the product mass, were placed inside the environment chamber. Airflow was monitored with an anemometer and kept constant at around 4 $m^3 s^{-1} m^{-2}$. Air temperature and relative humidity were monitored using a psychrometer installed near the sample trays.

During drying, these trays were periodically weighed. Hygroscopic equilibrium was deemed to have been reached when the mass variation of the containers remained approximately constant over three consecutive weighings (two replicates).

To determine the moisture ratio (MR) of the beans drying under the different conditions, the following expression was used:

$$MR = \frac{U^* - U_e^*}{U_i^* - U_e^*} \quad [8]$$

As a control for non-shrinkage, eight beans, wrapped in a fabric permeable to air were individually placed inside each tray.

The volume of each grain (V_g), considered to be a sphere, was obtained using the average value of the three orthogonal axes (a, b and c, in mm, Figure 1) as the diameter, as proposed by Mohsenin (1986). This measurement was taken for eight beans over the drying process, using a digital calliper. The volume was calculated using the following expression:

$$V_g = \frac{\pi \cdot a \cdot b \cdot c}{18} \quad [9]$$

Grain shrinkage (ψ_g) during drying was determined as the relationship between grain volume (V_g) at each time point and the initial volume (V_o) using the following expression:

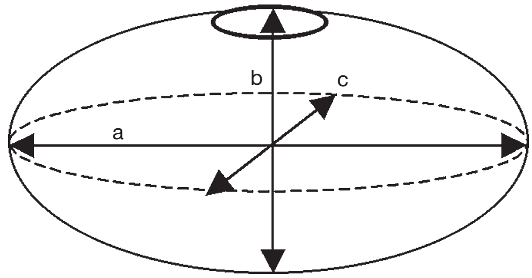

Figure 1. Schematic diagram of a bean grain showing the characteristic dimensions.

$$\psi_g = \frac{V_g}{V_o} \quad [10]$$

The liquid diffusion model (employing an approximation of eight terms [see Eq. 5]) was used with the experimental drying data, either taking into account grain shrinkage or neglecting it. The number of terms in the model was established when the variation of the diffusion coefficient was less than 0.1×10^{-13} $m^2 s^{-1}$.

The experimental data were interpreted by non-linear regression analysis using the Quasi-Newton method and employing STATISTICA 6.0® software (http://www.statsoftinc.com). The validity of the models with and without grain shrinkage was tested based on the significance of the regression coefficients (Student t test, $P = 0.01$), the mean relative error (MRE), the standard error of estimation (SEE), and the determination coefficient (R^2). Residual distribution plots were constructed to evaluate the fitting quality. The mean relative error and the standard error of estimation for the models with and without grain shrinkage were calculated according to the following expressions:

$$MRE = \frac{100}{n} \sum \frac{\left| Y - \hat{Y} \right|}{Y} \quad [11]$$

$$SEE = \sqrt{\frac{\sum \left(Y - \hat{Y} \right)^2}{GLR}} \quad [12]$$

where n is the number of experimental observations, Y the experimental value, $\hat{Y}$ the predicted value, and GLR the degrees of freedom of the model (number of observations minus the number of variables in the model).

To compare the diffusion models (as adjusted by [Eq. 5]) with and without grain shrinkage under each drying condition, the values estimated by the models were plotted. Linear regression analysis was then performed, with the straight line passing through the origin. The consistency of each model was assessed by determining the coefficient of determination (R^2) and the agreement expressed by the «d» indices proposed by Willmott *et al*. (1985). The «d» index indicates the degree of accuracy between the values estimated by the models, i.e., the closer to 1, the smaller the difference between the models. The «d» index is represented by the following expression:

$$d = 1 - \left[\frac{\sum \left(Y_e - Y_o \right)^2}{\sum \left(\left| Y_e - \overline{Y}_o \right| + \left| Y_o - \overline{Y}_o \right| \right)^2} \right] \quad [13]$$

where d is the agreement index, Y_e is the i[th] value estimated by model 1, Y_o is the i[th] value estimated by model 2, and $\overline{Y}_o$ the average of the values estimated by model 2.

Results

Figure 2 shows the experimental grain shrinkage values obtained during drying under the different conditions. Volume reduction varied between 18.9 and 35.2%, with the moisture content changing from 0.92 (kg water/kg dry matter) until the equilibrium moisture content specific for each set of conditions was reached.

Table 1 shows the mean diffusion coefficients, the relative and estimated mean errors, and the coefficients of determination for the liquid diffusion model with and without grain shrinkage under the drying different conditions.

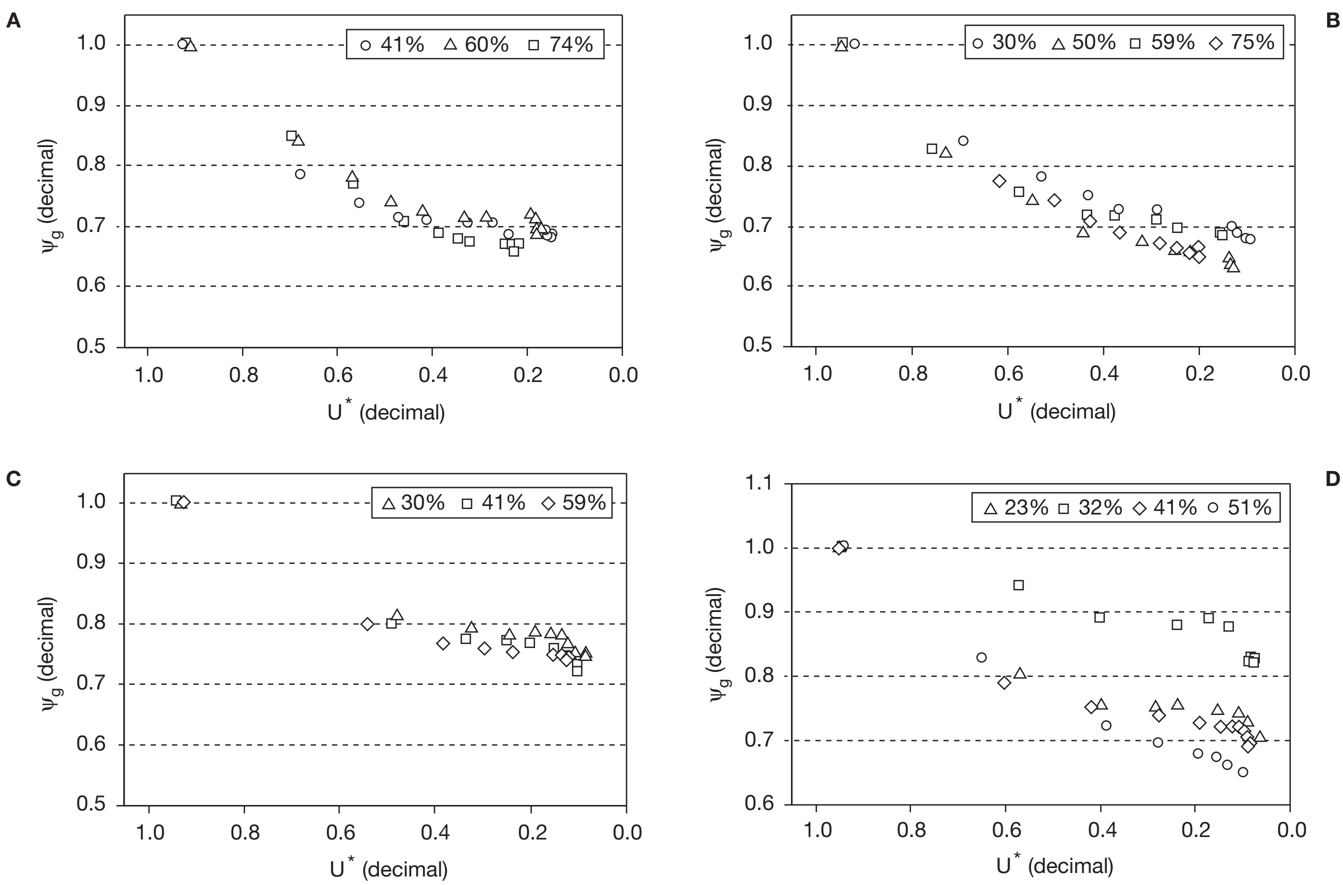

Figure 2. Experimental shrinkage (ψ_g) values as a function of moisture content (U*) for fourteen combinations of temperature (A: 25°C; B: 35°C; C: 45°C; D: 55°C) and relative humidity (23%, 30%, 32%, 41%, 50%, 51%, 59%, 60%, 74% and 75%).

Table 1. Coefficients of diffusion, mean relative error (MRE), standard error of estimation (SEE), and determination coefficient (R^2) for the liquid diffusion model with and without grain shrinkage (ψ_g), during the drying of beans under different conditions of temperature (T) and relative humidity (RH).

Air condition		R^2 (%)		SEE		MRE (%)		Coefficient of diffusion $\times 10^{-10}$ (m^2 s^{-1})	
T (°C)	RH (%)	With ψ_g	Without ψ_g	With ψ_g	Without ψ_g	With ψ_g	Without ψ_g	With ψ_g	Without ψ_g
25	41	98.69	98.51	0.0054	0.0062	26.84	25.78	1.67*	2.10*
	60	98.12	97.81	0.0077	0.0090	149.26	156.38	1.86*	2.28*
	74	99.42	99.30	0.0019	0.0023	49.77	49.73	2.21*	2.77*
35	30	99.45	99.45	0.0020	0.0021	47.07	46.23	3.27*	3.96*
	50	99.46	99.40	0.0019	0.0021	22.73	20.72	3.16*	4.03*
	59	99.29	99.15	0.0026	0.0032	23.48	21.10	3.22*	4.00*
	75	99.39	99.25	0.0019	0.0023	43.08	46.49	3.79*	4.81*
45	30	99.47	99.46	0.0014	0.0015	41.11	40.04	7.08*	8.26*
	41	99.38	99.36	0.0018	0.0019	26.47	25.07	7.43*	8.82*
	59	99.29	99.22	0.0022	0.0024	33.44	34.51	6.23*	7.43*
55	23	99.42	99.36	0.0018	0.0020	28.13	28.16	9.11*	10.84*
	32	99.44	99.42	0.0018	0.0019	48.67	48.80	8.65*	9.33*
	41	99.32	99.23	0.0022	0.0024	20.29	21.90	8.51*	10.38*
	51	99.40	99.32	0.0020	0.0023	39.73	38.57	5.72*	7.16*

* Significant at $P < 0.05$ (Student t test).

The SEE were small, confirming the validity of both models. However, the relative mean errors for both were > 10%, a less satisfactory result. Therefore, neither model appeared to be better than the other: these variables were insufficient to distinguish between the liquid diffusion models with and without grain shrinkage under the different drying conditions. Thus, «d» index values were calculated (Table 2).

Figure 3 shows the experimental moisture ratio values compared to values estimated by the liquid diffusion model for drying beans, taking into account the grain shrinkage under the different conditions of tem-

Table 2. «d»[1] indices used to compare the liquid diffusion models with and without shrinkage, the coefficients of determination (R^2), and the linear regressions (z), during drying under different temperatures (T) and relative humidities (RH)

T (°C)	RH (%)	d	R^2 (%)	Equation (z)
25	41	0.99997	99.99	z = 0.9971x
	60	0.99994	99.98	z = 0.9961x
	74	0.99988	99.96	z = 0.9931x
35	30	0.99996	99.99	z = 0.9967x
	50	0.99989	99.97	z = 0.9937x
	59	0.99994	99.98	z = 0.9956x
	75	0.99991	99.97	z = 0.9953x
45	30	0.99999	100	z = 0.9988x
	41	0.99999	100	z = 0.9988x
	59	0.99998	99.99	z = 0.9984x
55	23	0.99991	99.97	z = 0.9956x
	32	0.99998	100	z = 0.9982x
	41	0.99997	99.99	z = 0.9972x
	51	0.99990	99.97	z = 0.9939x

[1] Based on Willmott *et al.* (1985), Eq. [13].

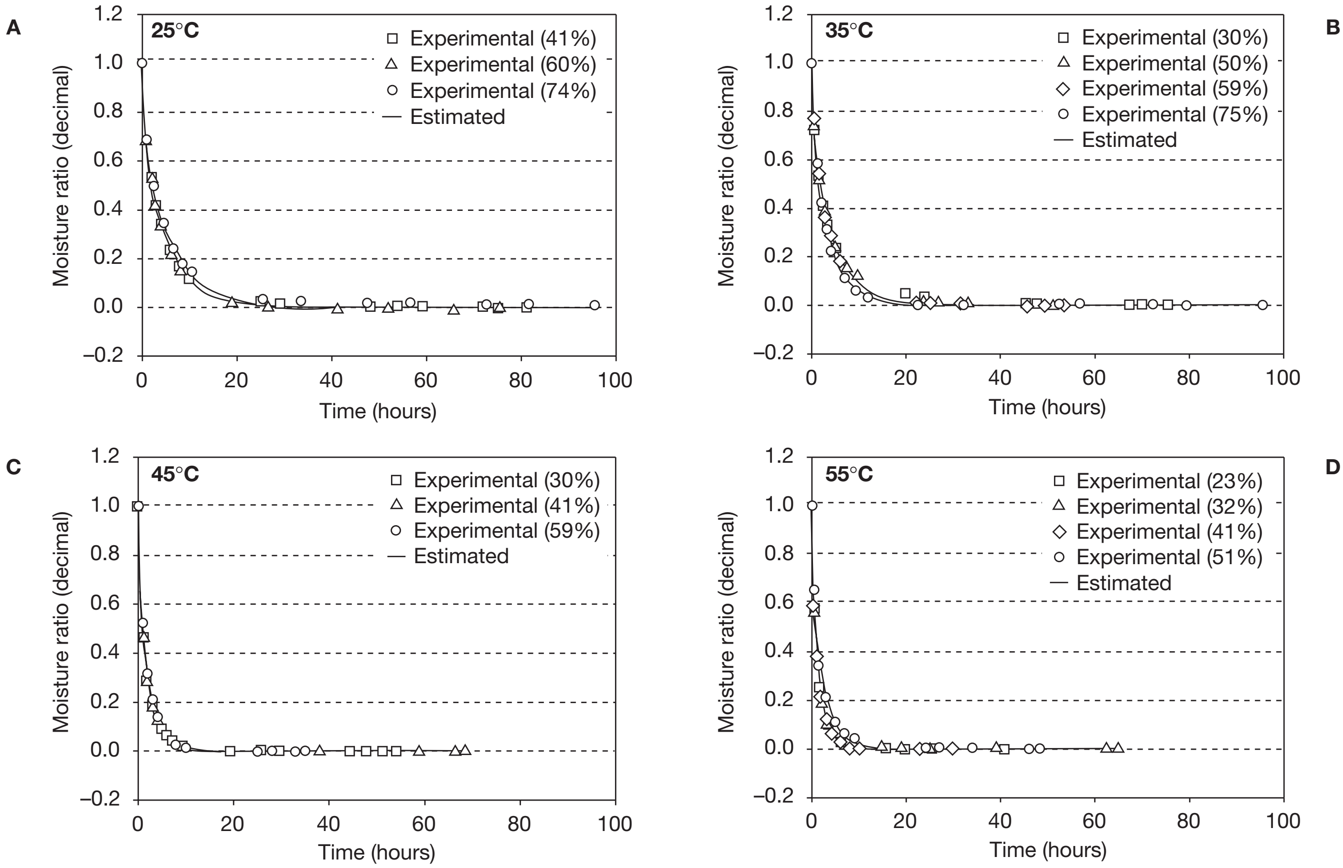

Figure 3. Experimental moisture ratio values and those estimated by the liquid diffusion model for the drying of beans, taking into account grain shrinkage under different temperatures (25°C, 35°C, 45°C and 55°C) and relative humidities (23%, 30%, 32%, 41%, 50%, 51%, 59%, 60%, 74% and 75%).

perature and relative humidity. The correspondence between the experimental and estimated values show that the liquid diffusion model without grain shrinkage appropriately describes the phenomenon studied.

The dependence of the diffusion coefficient on the drying temperature can be described by an Arrhenius-type relationship (Fig. 4). The coefficient of diffusion

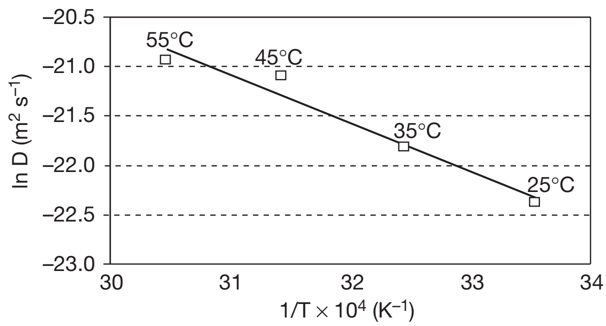

Figure 4. Arrhenius-type relationship between effective moisture diffusivity and temperature.

increased with the drying temperature. The slope [Eq. 6] provides the relationship E_a/R, while its intersection with the «y» axis indicates the value of D_o. Thus, the diffusion coefficient can be calculated as follows:

$$D = 3.02 \times 10^{-3} \exp\left(-\frac{40.08}{R \times T_a}\right)$$

where E_a in [Eq. 6] was 40.08 kJ mol^{-1} for temperatures ranging from 35ºC to 55°C.

The Arrhenius factor D_o is a constant equivalent to the diffusivity at infinitely high temperatures, and has the value 3.0×10^{-3} in drying beans. At this magnitude, this value can be neglected.

Discussion

When analysing grain shrinkage, Afonso Júnior *et al.* (2004) reported a reduction of 39% in the volume of coffee berries, with a reduction in their moisture content of 2.27 to 0.11 (kg water/kg dry matter). Ramos *et al.* (2005) observed a reduction of 35% of the radius of grapes during the drying process and indicated that the significant variation in the shrinkage of different products should be included in mass transfer models.

Table 1 shows that the liquid diffusion model (with or without grain shrinkage) satisfactorily describes the drying of the beans for all the air conditions studied. The regression coefficient was significant ($P < 0.05$) according to the Student t test, and the coefficient of determination was > 97.8%.

Table 1 also shows that the lowest drying air temperatures offer more internal resistance to water transport. Thus, a higher air temperature leads to increased diffusion coefficients and a greater outward movement of moisture (Babalis and Belessiotis, 2004; Sharma and Prasad, 2004).

During drying, the diffusion coefficients ranged between 1.67×10^{-10} and 9.11×10^{-10} m^2 s^{-1} when taking into account grain shrinkage, and between 2.10×10^{-10} and 10.84×10^{-10} m^2 s^{-1} (Table 1) when shrinkage was not taken into account. The calculated diffusion coefficients agree with those reported by other authors: Madamba *et al.* (1996) reported values of 10^{-9} to 10^{-11} m^2 s^{-1} for many products, while Doymaz (2005) indicated values of 4.27×10^{-10} to 1.30×10^{-9} m s^{-1} for temperatures between 50ºC and 70°C.

The liquid diffusion theory assumes that there is no influence of capillarity, neglects the effects of mass and energy transfer from one body to another (given the difficulty of quantifying these multiple effects on the product mass), and considers that thermal equilibrium with the air is reached instantaneously; this could lead to the discrepancies seen between the experimental and model-predicted results.

Table 2 shows a high «d» index (> 0.999) for all the temperature and relative humidity combinations, and excellent agreement between the values estimated by both models. The simple linear regression for the humidity ratio estimated by the models obtained high R^2 values (> 99.96%). This confirms the marked similarity and correlation between the liquid diffusion models that either take into account the volumetric contraction of the grains or neglect it. Carmo and Lima (2004) studied the drying of lentils using a liquid diffusion model that took shrinkage into account, and found it to predict experimental data very well.

Thermodynamically, E_a is the ease with which water molecules overcome the energy barrier when migrating within the product. In the drying processes, the smaller the E_a, the greater the water diffusivity within the product. The E_a value found in this work agrees with those reported for a number of biological materials, e.g., (in kJ mol^{-1}) soybean 28.8-30, wheat kernels 54-70.2, starch gel 18.8-50, scallion 29.05-42.05, canned mushroom 23.89-31.45, *Zea mays indentata corn,* 29.56, and rice 36.4 (Becker and Sallans, 1955; Fish, 1958; Park *et al.*, 1996; Bróvia *et al.,* 1997; Doymaz and Pala, 2003; Ramesh, 2003). Zogzas *et al.* (1996) noted that the E_a for these agricultural products ranged between 12.7 and 110 kJ mol^{-1}.

Based on the present results it is concluded that: i) the liquid diffusion model satisfactorily represents the kinetics of drying beans under a number of air conditions; ii) based on the analysed statistical variables, grain shrinkage can be neglected: the correlation between the models taking this into account and neglecting it was strong; iii) the diffusion coefficient increases with temperature, with values of 67.00×10^{-10} to 10.84×10^{-10} m^2 s^{-1} seen for air temperatures ranging from 25ºC to 55°C and relative humidities of between 20 and 75%; iv) the relationship between the diffusion coefficient and temperature is described by the Arrhenius equation, with an activation energy for liquid diffusion of 40.08 kJ mol^{-1}.

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