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Effective Moisture Diffusivity Estimation from Drying Data

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Abstarct: Drying is a complicated process of simultaneous heat and mass transfer. A significant number of mechanisms for moisture transportation within the porous solids have been proposed. Unfortunately, none of these mechanisms can be said to prevail throughout the total drying process. It seems that it is not possible to suggest a standard method for the evaluation of moisture diffusivity. However, moisture diffusivity estimation from drying kinetics data seem to provide more accurate results. This article presents the same simplified methods for determination of the effective diffussion coefficient of agricultural materials from experimental drying kinetics data. Each particular method is briefly described and also discussed by using experimental drying data. From these methods, Lewis's Simplified Method can be proposed for routine engineering calculations, where diffusivity can be represented by a mean constant value. In cases where diffusivity is strongly depended on moisture content, slope, MQM and a proper numerical method may provide the best results.
Key words: Diffusivity, effective diffusivity, apparent diffusivity

Kuruma Verilerinden Yararlanılarak Efektif Nem Difüzyonunun Tahminlenmesi

Özet: Kuruma sürekli olarak ısı ve kütle iletimini içeren karmaşık bir işlemdir. Gözenekli yapıdaki katı bir cismin içinde oluşan nem taşınması işleminde çok sayıda mekanizma etkilidir. Ancak, bu mekanizmaların hiç biri kuruma sürecinin başından sonuna kadar aynı etkinlikte yer almazlar. Bu nedenle nem difüzyonu işlemini tanımlayan standart bir yöntem önerilemez. Buna rağmen, materyalin kurumasıyla ilgili kinetik verilerden yararlanılarak nem difüzyonunun tahmininin en doğru sonuçları verdiği söylenebilir. Bu makalede nem difüzyonunun tahminlenmesinde kullanılabilecek yöntemler açıklanmış ve örnekler ile bu yöntemler karşılaştırılmıştır. Bu yöntemler içinde, difüzyonun değişmediği söylenebilen durumlarda, Lewis'in basitleştirilmiş yönteminin genel mühendislik işlemleri için yeterli olduğu kabul edilebilir. Difüzyonun materyalin nem içeriğine çok bağlı olduğu durumlarda ise eğim, MQM ya da uygun bir nümerik yöntem en iyi sonucu verecektir.
Anahtar kelimeler: Difüzyon, efektif difüzyon

INRODUCTION

Drying is a complicated process of simultaneous heat and mass transfer. A significant number of mechanisms for moisture transportation within the porous solids have been proposed to explain the drying or rehydration phenomena such as molecular diffusion, capillary motion, liquid diffusion through solid pores, vapour diffusion in air filled pores, vaporization–condensation sequence flow, hydrodynamic flow, and change of volume, shape and texture of material.

On the other hand none of these mechanisms can be said to prevail throughout the total drying process.

Drying of agricultural materials can be divided into constant and falling rate periods, the former is rarely observed in the drying of this kind of materials. In this case, the controlling mechanism of moisture transportation would be that of internal movement, so moisture is considered to move internally by diffusion and externally by forced convection. It can be said that, diffusivity is the transfer rate of water molecules in porous bodies to different directions in a unit time by random molecular motion.

In recent years, modeling of drying process has become an attractive target for most researchers in the field. Optimization of such processes inevitably incorporates precise models, so that, moisture and temperature-time profiles can be reasonably predicted within the porous solid bodies. This means that, transport properties like, diffusion, mass and heat transfer coefficients, as well as, thermal conductivity, must be accurately estimated. Additional properties like equilibrium moisture content, shrinkage, bulk density, specific volume and porosity are also required.

The diffusivity of water in agricultural products is an important property which is useful in prediction and engineering analysis of various mass transfer operations, such as drying, rehydration, and storage. Due to the complex chemical composition and wide diversity of physical structure of agricultural materials, reliable data on diffusivity of water in agricultural materials are not available for many of them. Modern drying process technologies of agricultural products are more complex, and more precise data on agricultural material are needed for accurate analysis, design and control of industrial driers (Zogzas *et al.*, 1994). Thus, experimental measurement of the diffusivity becomes necessary for many marketable dried agricultural products (Saravacos and Raouzeos, 1984). The diffusivity of moisture depends not only on the nature of the agricultural product, but also on moisture content and the temperature (Henderson and Papis, 1961; Saravacos, 1984, Mujumdar and Devehastin, 2000). Depending upon the physical structure of the material and the drying condition, water can be transported by a combination of mechanisms as mentioned before (Saravacos and Raouzeos, 1984; Sablani *et al.*, 2000).

When different transport mechanisms occur, it is difficult to separate individual mechanism, and the rate of moisture movement is described by an effective diffusivity, D_{eff}, irrespective of which mechanism is really involved in moisture movement (Sablani *et al.*, 2000; Katekawa and Silva, 2006). Thus, the experimentally estimated transport property represents the effective or apparent diffusivity of moisture in the agricultural product.

SOME METHODS to ESTIMATE the EFFECTIVE DIFFUSIVITIES of AGRICULTURAL MATERIALS

Although most of the diffusivity estimation methods are based on Fick's laws of diffusion, there are significant differences in the way of applying these laws on experimental data as well as in the kind of experiments used. It is clear that there is no standard method of evaluating the moisture diffusivity.

Fick's second law of diffusion is often used to describe a moisture diffusion process (Andrieu and Stamatopoulos, 1984; Zogzas *et al.*, 1994; Liu *et al.* 1998; Sablani *et al.*, 2000; Alvarez and Legues, 1986):

$$\frac{\partial M}{\partial t} = D_{eff} \nabla^2 M \qquad (1)$$

where, M is the local moisture content (dry basis), t is the time (h) and D_{eff} is the moisture diffusivity (m^2h^{-1}). In engineering applications, one-directional diffusion is a good approximation for most practical systems (Saravacos, 1986). Thus, in most situations, the food product is assumed as one-dimensional. The solutions of the Fickian equation in such conditions for different geometries has been presented by many authors (Papis and Henderson, 1961; Crank, 1975; Tang and Sokhansanj, 1993).

To solve the Eqn (1) following assumption must be applicable: (i) material has the constant diffusivity, (ii) material has the uniform moisture distribution, (iii) surface moisture of the material is equal to the equilibrium moisture content.

If the diffusivity is assumed constant within a certain moisture range, integration of Eqn (1) gives the following solutions for infinite slap [Eqn (2.1)], infinite cylinder [Eqn (2.2)] and sphere [Eqn (2.3)] geometry drying both surfaces (Zogzas and Maroulis, 1996):

Infinite slap: $$M_R = \frac{M_t - M_e}{M_{cr} - M_e} = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}exp\left[-\frac{(2n+1)^2\pi^2 D_{eff}t}{L^2}\right] \quad (2.1)$$

Infinite cylinder: $$M_R = \frac{M_t - M_e}{M_{cr} - M_e} = \sum_{n=1}^{\infty}\frac{4}{b_n^{\,2}}exp(-b_n^{\,2}\frac{D_{eff}t}{r_s^{\,2}}) \quad (2.2)$$

Sphere: $$M_R = \frac{M_t - M_e}{M_{cr} - M_e} = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}exp(-n^2\frac{D_{eff}t}{r_k^{\,2}}) \quad (2.3)$$

where, M_R is dimensionless moisture ratio; M_t is the moisture content at any time in falling rate period of drying ($kgkg^{-1}$); M_e is dynamic equilibrium moisture content for the air conditions existing in the drying chamber ($kgkg^{-1}$); M_{cr} is critic initial moisture content at the beginning of the falling rate period (kg/kg); t is time (h); D_{eff} is effective diffusivity (m^2h^{-1}); L is half thickness of the slap (m); r_s and r_k are radius of cylinder and sphere, respectively, n is positive integer represents the number of the terms in the summation series and b_n is characteristic root of first kind and zero order Bessel functions (b_1=2.4048).

The number of terms n, necessary for calculation of the moisture content from Eqn (2) with an accuracy of 1% for the beginning of drying is n=20. When using only one term of the series for the beginning of the drying process, the error reaches 19% (Efremov and Kudra, 2005). If drying occurs only one surface of the slab, the thickness L in Eqn (2) must be substituted by 2L.

Equation (2) can be simplified with an acceptable error for long drying times to drying kinetics prediction for the fist stage of falling rate period of drying as in the following (Papis and Henderson, 1961; Bagnoli *et al.*, 1973; Zogzas and Maroulis, 1996).

$$M_R = \frac{8}{\pi^2}exp(-\pi^2 Fo) \quad (3)$$

where Fo denotes the Fourier number *(Fo= $D_{eff}\,t/L^2$).*

From Eqn (3), it is clear that at the beginning of drying (t=0) M_R is equal 0.81 instead of unity, so this equation can be used only for approximate calculations, especially for short drying times (Efremov and Kudra, 2004; Efremov, 2006).

The effective diffusion coefficient D_{eff} may be calculated from Eqn (3) where, the term $8/\pi^2$ is considered equal to unity [Eqn (4)] as in the following [Eqn (5)], knowing the experimental values of M_R (Efremov and Kudra, 2004).

$$M_R = exp(-\pi^2 Fo) \quad (4)$$

$$D_{eff} = \frac{L^2}{\pi^2 t}(-ln\, M_R) \quad (5)$$

Efremov and Kudra (2005) pointed out that the results of this simplified Eqn (4) gives significant deviations of experiments from the model predictions in a regular regime (Fo>0.04), reaching 23.4% for a plate and 64.5% for a sphere, and they proposed to following equation to reduce these deviations,

$$M_R = exp(-\pi^2 Fo^a) \quad (6)$$

where, a is the correction factor, for a plane sheet a=0.91 and for a sphere a=0.83 with the maximum relative deviation of ±12% and ±17%, respectively.

D_{eff} may be calculated from Eqn (6) as in the following Eqn (7).

$$D_{eff} = \frac{L^2}{\pi^{2/a}\, t}(-ln\, M_R)^{a^{-1}} \quad (7)$$

Another simplified approach to predict drying kinetics for the first falling rate period of drying, has been introduced by Lewis in 1921, is called thin layer equation (Henderson and Pabis, 1961; Pabis and Henderson, 1961):

$$\frac{\partial M}{a\,\partial t} = -K(M - M_e) \quad (8)$$

where, K is an empirical constant called drying constant (h^{-1}) and a is constant, dependent on geometric shape of the material.

Obviously, the drying constant K, can be considered a combination of transport properties encountered during drying, like moisture diffusivity, thermal conductivity, mass and heat transfer coefficients (Zogzas and Maroulis, 1996). The use of such a simplified equation, like the thin layer one, is extremely useful and time saving in process design situations.

The following Lewis's well known drying equation is derived by integrating the thin layer Eqn (8) between initial and mean moisture content at time t.

$$M_R = exp(-K\,t) \tag{9}$$

Comparing Eqn (4) and (9), drying constant, K, can be related to moisture diffusivity by the relation

$$K = \frac{\pi^2 D_{eff}}{L^2} \tag{10}$$

$$D_{eff} = K\left(\frac{L}{\pi}\right)^2 \tag{11}$$

Equation (10) relates the empirical drying constant K, to the theoretical property of moisture diffusivity D_{eff} (Zogzas and Maroulis, 1996; Doymaz *et al.*, 2004).

According to the Lewis's simplified Eqn (9), the effective diffusivity (D_{eff}) can be estimated from the slope of a semilogarithmic diagram of moisture ratio (M_R), which is obtained from the experimental data, versus time (t) (Saravacos, 1986; Zogzas *et al.*, 1994). This plot is a straight line over the first falling drying period, but it deviates at late drying stages (Bagnoli *et al.*, 1973; Zogzas *et al.*, 1994). Curve fitting procedure is performed to estimate the best mathematical model to define the experimental drying curve and determine their slope. The slope of this straight line is considered equal to the quantity ($\pi^2 D_{eff}/L^2$), from which diffusivity is determined. As an example, the moisture content data obtained at 60°C drying air temperature of bay leaves were converted to the dimensionless moisture ratio expression, M_R, and then experimental drying curves (log M_R vs t) of bay leaves were plotted as shown in Fig. 1 (Yagcioglu *et al.*, 2001). Obviously, the method described above, cannot be used in cases where diffusivity depends strongly on moisture content. Alternatively, zone method can be used that is based on splitting the entire kinetic curve into several zones over, and the Lewis's simplified equation can be applied to assumption of constant diffusivity is an acceptable approximation (Fig. 2).

However an alternative procedure is described by Bagnoli *et al.*, (1973) and by Saravacos and Raouzeos (1984), in order to compensate for the case of moisture dependent diffusivity. This method involves the comparison of the slope of experimental drying curve log M_R vs t to the slope of theoretical diffusion curve log M_R vs Fourier number (F_o). First, the theoretical moisture ratios M_R are evaluated numerically for a range of Fourier numbers F_o. Then, the same ratios M_R are evaluated using experimental data. Both curves of experimental and theoretical M_R are plotted vs time and Fourier number, respectively on a semilogarithmic diagram (Zogzas *et al.*, 1994).

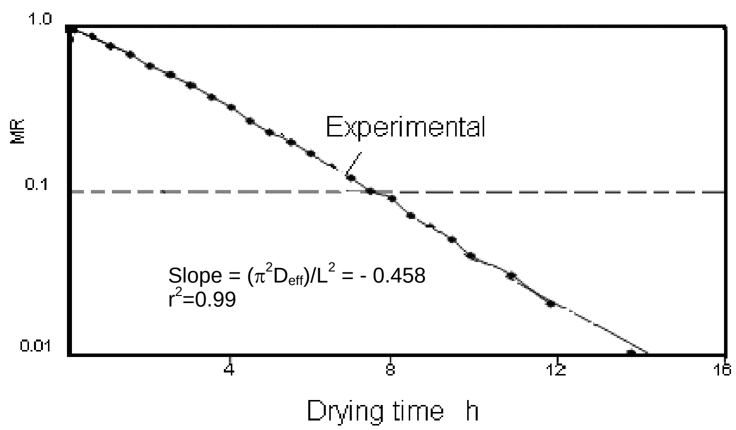

Figure 1. Log M_R vs t curve of bay leaves at 60 °C

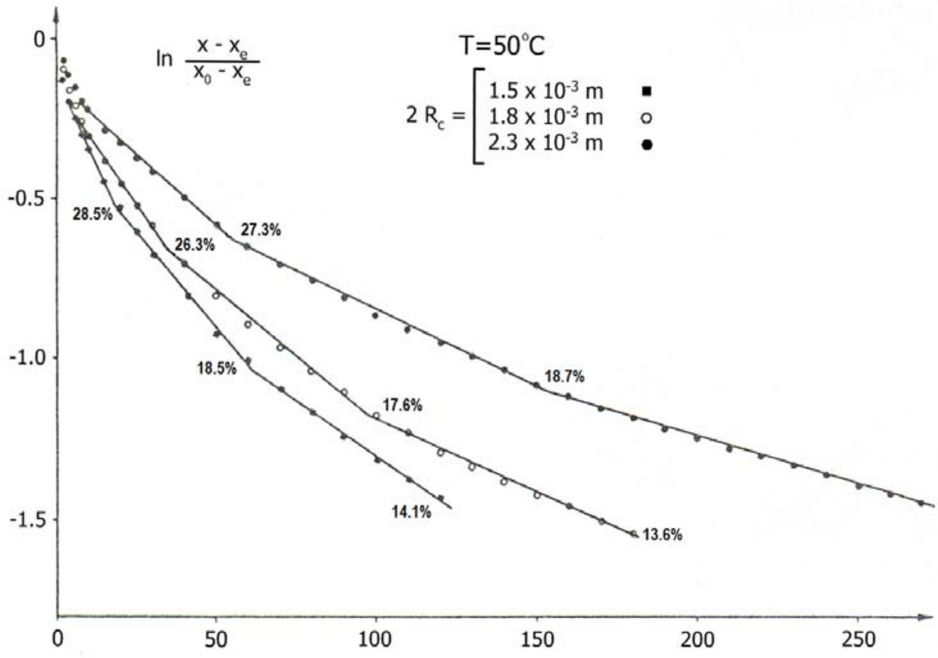

Figure 2. Kinetic curve zones of spaghetti as a function of diameters (Andrieu and Stamatopoulos, 1984).

As an example, the moisture content data obtained at 60°C drying air temperature of bay leaves were converted to the dimensionless moisture ratio expression, M_R, and then experimental drying curves (log M_R vs t) of bay leaves and theoretical (log M_R, vs Fourier number), were plotted as shown in Fig. 3 (Yagcioglu *et al.*, 2001)

The slopes of experimental and theoretical curves can be determined by numerical differentiation. Then the effective diffusivity of moisture at a certain moisture content can be estimated from the following equation (Bagnoli *et al.*, 1973; Saravacos and Raouzeos, 1984; Saravacos, 1986; and Sablani *et al.*, 2000).

$$D_{eff} = \frac{\frac{dM_R}{dt}}{\frac{dM_R}{dFo}} L^2 \qquad (12)$$

Since there is a moisture content value, M_t, which correspond to the specified moisture ratio, M_{Rt}, D_{eff} can be found as a function of moisture content, by applying Eqn (12) over the range of M_R.

In most cases, effective moisture diffusion coefficient is considered as constant even though noticeable discrepancies in kinetic data occur between experiments and analytical solutions in the form of the Fourier series. Alternatively, zone method can be used that is based on splitting the entire kinetic curve into several zones over which the assumption of constant diffusivity is an acceptable approximation. This limitation can be overcome when applying the following equation that was obtained by modifying the quasi-stationary method, MQM, in order to describe kinetics of mass transfer (Efremov and Kudra, 2005).

$$M_R = \frac{1}{1 + (\frac{t}{\sigma})^m} \qquad (13)$$

where, σ is characteristic time (s, h) which assumes a constant value for the given drying condition, and m is the index of hydrodynamic intensity (dimensionless). The index of hydrodynamic intensity, m, and characteristic time, σ are generated from the experimental values of M_R

Combining Eqn (7) and (13) gives Eqn (14).

$$D_{eff} = \frac{L^2}{\pi^{2/a} t} \left\{ ln\left[1 + \left(\frac{t}{\sigma} \right)^m \right] \right\}^{a-1} \qquad (14)$$

The advantage of Eqn (14) is the continuous function for the time depended effective moisture diffusivity, as opposed to discrete values obtained when using the zone method. If m>1, the effective diffusion coefficient at beginning of drying process increases with time and then decreases, and if m<1, the effective diffusion coefficient decreases with time.

To confirm the most suitable calculation method of effective diffusivity of the material, dimensionless moisture ratio values, M_R, calculated from the models by using estimated effective diffusivities, $D_{eff,}$ and than statistical analysis is achieved to compare them with experimental data. The coefficient of correlation, r, is one of the primary criterion for selecting the best model to estimate the effective diffusivity. In addition to r, mean bias error, E_{MB}, and root mean square error, E_{RMS}, are used to determine the quality of the fit. The higher the values of the r, and lowest values of the E_{MB} and E_{RMS}, the better to goodness of the fit.

As an example, the results of statistical analysis of experimental and calculated M_R values of olive dried at different air temperatures in Table 1 (Demir *et al.*, 2007).

From Table 1 it is clear that the D_{eff} values calculated from Lewis's simplified model have the better fit than the others to experimental M_R values.

An addition of statistical error analysis, to confirm the most suitable calculation method of effective diffusivity, moisture ratio values, M_R, calculated from the models by using estimated effective diffusivities, D_{eff}, and than M_R vs. time curves can be plotted and compared by the experimental plots.

As an example, the comparison of experimental dimensionless moisture ratios, M_R-t curve, with the calculated values of M_R-t curves of bay leaves at Fig. 4 (Demir *et al.*, 2004).

According to the plots at Fig. 4 it is clear that the calculated based Lewis model has better fit to experimental M_R-t curve than the others for this example.

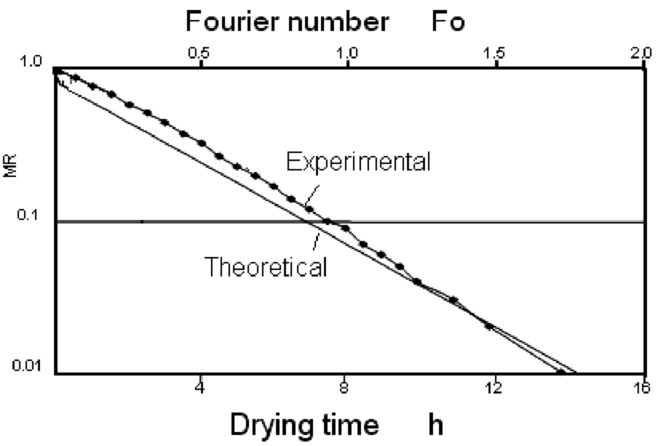

Figure 3. Experimental and theoretical drying curves of bay leaves at 60°C

Table 1. Comparison of r, E_{MB} and E_{RMS} values of M_R values, which were estimated by different D_{eff} calculation methods, with experimental M_R values of olive

	50°C			60°C			70°C		
	MQM	Lewis	Slope	MQM	Lewis	Slope	MQM	Lewis	Slope
E_{MB}	0.0157	0.0042	-0.045	0.016	0.005	-0.040	0.0093	-0.0015	-0.0106
E_{RMS}	0.0413	0.0194	0.062	0.0395	0.0148	0.0585	0.026	0.0066	0.0209
r	0.98	0.99	0.98	0.99	0.99	0.98	0.99	0.997	0.99

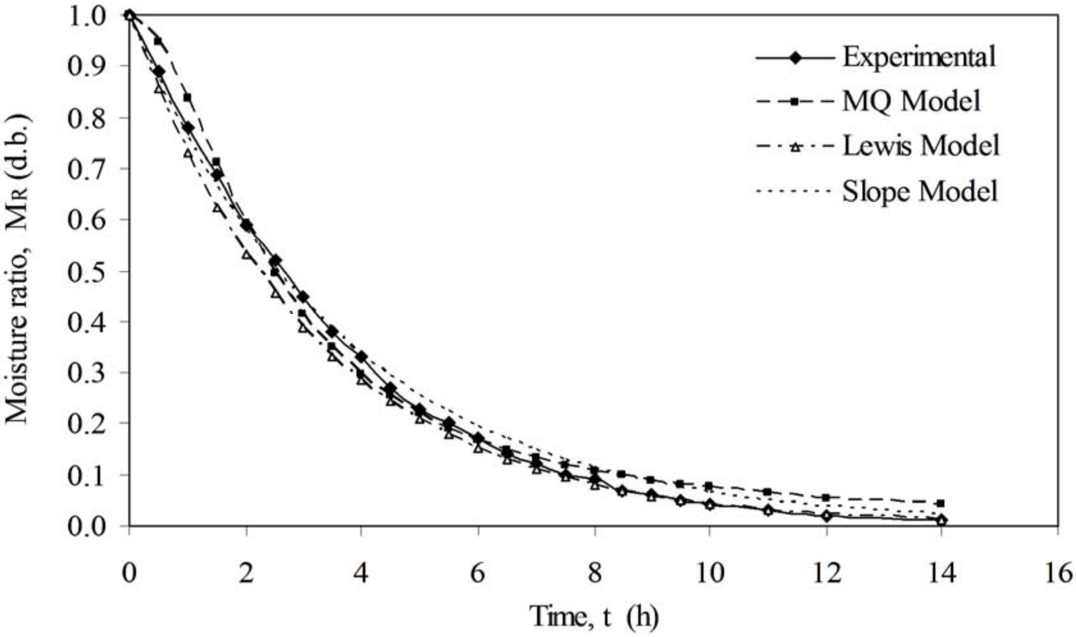

Figure 4. Comparison of, based on the predicted values of D_{eff} from Lewis, MQM and slope model at 40°C of bay leaves.

CONCLUSION

It seems that it is not possible to suggest a standard method for the evaluation of moisture diffusivity. The researchers have to chose among a variety of methods and techniques to find the one which is best suited to the particular needs of their experiments.

However, the drying methods seem to provide more accurate results, since the mass boundary layer formation can be kept to a minimum, thus reducing significantly the resistance to mass transfer at the interference (Zogzas *et al.*, 1994). From these methods, Lewis's simplified method can be proposed for routine engineering calculations, where diffusivity can be represented by a mean constant value. In cases where diffusivity is strongly depended on moisture content, slope, MQM and a proper numerical method may provide the best results.

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