Effective Moisture Diffusivity Estimation from Drying Data

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Abstract: 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 diffusion 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

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).

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):

\[
\frac{\partial M}{\partial t} = D_{\text{eff}} \nabla^2 M
\]  

where, \( M \) is the local moisture content (dry basis), \( t \) is the time (h) and \( D_{\text{eff}} \) is the moisture diffusivity (m²h⁻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 slab [Eqn (2.1)], infinite cylinder [Eqn (2.2)] and sphere [Eqn (2.3)] geometry drying both surfaces (Zogzas and Maroulis, 1996):

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

\[
M_R = \frac{M_i - M_e}{M_{cr} - M_e} = \frac{4}{\pi^2} \sum_{n=1}^{\infty} \frac{4}{b_n^2} \exp\left(-\frac{b_n^2 \pi^2 D_{eff} t}{r_s^2}\right)
\]  
(2.2)

\[
M_R = \frac{M_i - M_e}{M_{cr} - M_e} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(-\frac{n^2 \pi^2 D_{eff} t}{r_k^2}\right)
\]  
(2.3)

where, \(M_R\) is dimensionless moisture ratio; \(M_i\) is the moisture content at any time in falling rate period of drying (kg/kg\(^{-1}\)); \(M_e\) is dynamic equilibrium moisture content for the air conditions existing in the drying chamber (kg/kg\(^{-1}\)); \(M_c\) is critical initial moisture content at the beginning of the falling rate period (kg/kg\(^{-1}\)); \(t\) is time (h); \(D_{eff}\) is effective diffusivity (m\(^2\)h\(^{-1}\)); \(L\) 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 first stage of falling rate period of drying as in the following (Papis and Henderson, 1961; Bagnoli et al., 1973; Zogzas and Maroulis, 1996).

\[
M_R = \frac{8}{\pi^2} \exp(-\pi^2 Fo) 
\]  
(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).

\[
D_{eff} = \frac{L^2}{\pi^2 t} (-\ln M_R) 
\]  
(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,

\[
M_R = \exp(-\pi^2 Fo^a) 
\]  
(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).

\[
D_{eff} = \frac{L^2}{\pi^2 t} (-\ln M_R)^{a^{-1}} 
\]  
(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):

\[
\frac{\partial M}{\partial t} = -K(M-M_e) 
\]  
(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.

\[ M_R = \exp(-K \cdot t) \]  

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

\[ K = \frac{\pi^2 D_{eff}}{L^2} \]  

\[ D_{eff} = K \left( \frac{L}{\pi} \right)^2 \]  

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

\[ \text{Slope} = \left( \pi^2 D_{eff}/L^2 \right) = 0.458 \]

\[ r^2=0.99 \]
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).

$$D_{eff} = \left(\frac{dM_R}{dt}\right) \frac{L^2}{dM_R}$$  \hspace{2cm} (12)

Since there is a moisture content value, $M_t$, which correspond to the specified moisture ratio, $M_{R0}$, $D_{eff}$ 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).

$$M_R = \frac{l}{l + (\frac{t}{\sigma})^m}$$  \hspace{2cm} (13)

where, $\sigma$ 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, $\sigma$ are generated from the experimental values of $M_R$.

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

$$D_{eff} = \frac{L^2}{\pi^{\frac{3}{2}} t} \left\{ \ln \left[ 1 + \left( \frac{t}{\sigma} \right)^m \right] \right\}^{a^{-1}}$$  \hspace{2cm} (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, \( \dot{M}_R \), 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 \( \dot{M}_R \) 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](image)

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

<table>
<thead>
<tr>
<th></th>
<th>50°C</th>
<th>60°C</th>
<th>70°C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MQM</td>
<td>Lewis</td>
<td>Slope</td>
</tr>
<tr>
<td>( E_{MB} )</td>
<td>0.0157</td>
<td>0.0042</td>
<td>-0.045</td>
</tr>
<tr>
<td>( E_{RMS} )</td>
<td>0.0413</td>
<td>0.0194</td>
<td>0.062</td>
</tr>
<tr>
<td>( r )</td>
<td>0.98</td>
<td>0.99</td>
<td>0.98</td>
</tr>
</tbody>
</table>
CONCLUSION

It seems that it is not possible to suggest a standard method for the evaluation of moisture diffusivity. The researchers have to choose 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 dependent on moisture content, slope, MQM and a proper numerical method may provide the best results.

REFERENCES


