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Inversion of a numerical model to predict the effective moisture diffusivity of fruits during drying as a function of temperature and moisture content

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Abstract

Drying is one of the most important fruit and vegetable preservation method useful to remove water from fresh materials. This permits to avoid growth of bacteria, yeasts or molds. To better understand the heat and mass transfer process that take place during drying, numerical models can be used. The effective diffusivity determination is a crucial aspect for the development of drying models. The diffusivity is affected by the product temperature and moisture content. The aim of this study was to develop a method, based on the inversion of a finite element model, to estimate the moisture diffusivity of fruits during drying as a simultaneous function of temperature and moisture content. The research work was divided in three phases: i) experimental determination of the moisture concentration versus time in various fruits during the drying process; ii) development of a numerical heat and mass transfer model for the determination of moisture content versus time; iii) parameter estimation of moisture diffusivity as a function of temperature and moisture content, by minimizing the distance between numerical model and experimental results using a feasible optimization algorithm. The estimated moisture diffusivity coefficients are close to those reported in literature for the same fruits (banana and apple), but obtained by others approaches. The experimental and calculated moisture contents are in good agreement showing a determination coefficient $R^2 > 0.97$.

Keywords: fruit drying, inverse method, finite element model

INTRODUCTION

In order to reduce the trial and error effort involved in experimental test and to better understand the heat and mass transfer processes that take place during the fruit drying, several numerical models have been developed (Castro et al., 2018). To obtain suitable models, a good knowledge of heat and moisture transport properties of the involved material is essential. Furthermore, using numerical models, the physical properties may be described by time/space/temperature/moisture-dependent variables (Fabbri et al., 2014). However it is not easy to find in literature mass transfer food properties, compared to thermal ones, especially measured or calculated during the drying processes. As concerning the moisture diffusivity, this property strongly depends on temperature and on the moisture changes involved during the heat process (Guillard et al., 2003).

The main diffusivity estimation methods are based on Fick's laws, but there are significant differences in the implementation; there is no standard method of evaluating the moisture diffusivity in food (Zogzas et al., 1994). Nevertheless the most common method is the fitting of the experimental drying data in a logarithmic form; for a simple one-dimensional geometries, by assuming the first term approximation in the series solution, the diffusivity can be achieved from the relative slope (slope method)(Arranz et al., 2017). This simple method cannot be use to describe the diffusivity changes a simultaneous function of product temperature and moisture content. To do this, an Arrhenius type equation is commonly used to model the temperature effect, while the dependence of the moisture is introduced comparing the slopes of two curves at specific moisture ratio (Zogzas et al.1994).

In recent years, numerical method has become a quite used methodology to evaluate the moisture diffusivity during vegetable dryin (Castro et al., 2018). Generally the diffusivity is introduced in the numerical model through Fick's equations as a function of temperature or moisture content or any other material properties (Khan et al., 2017). The solution is found by fitting the numerical solution provided by the model to the measured data, by varying the model parameters. (Fabbri et al., 2014). Few research works are based on inversion of finite element (FE) models of drying fruit (da Silva et al., 2009, da Silva et al., 2010; Silva et al., 2012; Balzarini et al., 2018), while the majority reports the use of the finite difference methods.

The aim of this work was to determine the moisture diffusivity of apple and banana fruit during the drying process by using the inversion of a finite element model. Three drying temperature were considered (50, 60 and 70°C) to check the temperature and moisture content influence. The results were compared with those obtained by the classical slope method.

MATERIALS AND METHODS

1. Fruit samples and experimental drying

Ripe bananas (var. Cavendish) and apples (var. Golden Delicious) were purchased from a local market (Cesena, FC, Italy). The fruits were cut into slabs of 3 mm thickness, by using a manual slicing machine, transversally to the prevalent dimension; subsequently, cylindrical samples (diameters of 30 mm) were obtained by using a small hollow cutter. Twenty samples for each fruit type were used for the drying experiment. The initial moisture content of the samples was determinate by the oven drying method following the AOAC, 2000. A convective dryer was used to dry the fruit samples from the initial moisture content to the equilibrium moisture content between the samples and the drying air. Three different air temperatures (50, 60 and 70°C; air velocity: 0.2 m s⁻¹) were tested. The samples were placed in the dryer after 30 min from the turn on and was weighted before to start the experiment and after every 15 minutes, until a constant weight. Subsequently, the moisture content (dry basis) was calculated on the base of the initial moisture. At the same time intervals, sample thickness (*l*) and diameters (*d*) were measured by using a digital caliper (CDJB15 Borletti, Italy). The air and fruit (in the geometric centre) temperature was monitored by using K-type thermocouples (Chromel/Alumel; Tersid Came, Milano, Italy). The data were sampled at a frequency of 0.25 Hz, with a precision of 0.1°C, using the acquisition system PCI-6036E (National Instrument Corporate, Austin). All drying test were performed in triplicate.

2. Moisture diffusivity determination

2.1. Inverse method.

The diffusivity was calculated by the inversion of a finite element model. The work was divided in three phases: I) experimental determination of the moisture concentration (\bar{C}_{exp}) versus time during drying (section 1); II) development of a 2D-axisymmetric model of the moisture and heat transfer in the fruit during drying and numerical determination of the moisture content (\bar{C}_{num}) vs time; III) parameter estimation of moisture diffusivity, minimizing the distance between numerical model and experimental moisture content data.

Numerical model

The 2D-axisymmetric finite element model was developed using Comsol Multiphysics (COMSOL Inc., Burlington, MA). The geometric model replied the real dimensions (radius and height) and shapes of the samples used for the experimental test.

The mass transfer was governed by the 2nd Fick's law:

$$\frac{\partial C}{\partial t} = \nabla \cdot (D \nabla C) \quad (1)$$

where *C* (mol m⁻³) is the calculated moisture concentration at time *t* (s);

while the heat was transferred by conduction following the Fourier's equation:

$$C_p \rho \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) \quad (2)$$

where C_p (J K⁻¹kg⁻¹), k (W m⁻¹K⁻¹) and ρ (kg m⁻³) are the specific heat, thermal conductivity and density of the fruits, respectively (see table 1)

As concerning the boundary conditions, flux conditions were imposed on the interface between the fruit sample and the dryer air.

Mass flux

$$n \cdot (-D \nabla C) = N = h_m \left(\frac{P_\infty}{RT_\infty} - \frac{P_s}{RT_s} \right) \quad (3)$$

where N (molm⁻²s⁻¹) is the water molar flux, R (Jmol⁻¹K⁻¹) the universal gas constant and h_m (m s⁻¹) the mass transfer coefficient calculated on the basis of the well-known Chilton-Colburn analogy between the Nusselt number and the Sherwood number (Sh):

$$h_m = \frac{Sh D_a}{L} \quad (4)$$

$$Sh_t = 0.54(Gr_m Sc)^{1/4} \text{ for the top surface} \quad (5)$$

$$Sh_b = 0.27(Gr_m Sc)^{1/4} \text{ for the bottom surface} \quad (6)$$

$$Sc = \frac{\mu_a}{\rho_a D_a}: \text{Schmidt numbers} \quad (7)$$

$$Gr_m = \frac{gL^3 \rho_a (\rho_{as} - \rho_{a\infty})}{\mu_a^2}: \text{Grashof number for the mass flux} \quad (8)$$

$$\rho_{as} = X_s \rho_a: \text{density of humid air at the fruit surface (kg m}^{-3}\text{)} \quad (9)$$

$$\rho_{a\infty} = X_\infty \rho_a: \text{density of humid air far from the fruit surface (kg m}^{-3}\text{)} \quad (10)$$

The dried vapour pressure far from the product surface (P_∞), and the vapour pressure close to samples (P_s), were determined on the base of the vapour relative humidity in the dryer far from the product surface (RH_∞) and the water activity at the interface (aw), together with the corresponding temperature, via saturated vapour pressure $P_{sat}(T)$ given by Antoine's law:

$$P_{sat}(T) = \left[10^{\left(8.07131 - \frac{1730.63}{233.426 + T} \right) \frac{10^5}{760}} \right] \quad (11)$$

$$P_\infty = RH_\infty P_{sat}(T_\infty) \quad (12)$$

$$RH_\infty = \left(\frac{X_\infty P_{atm}}{0.622 P_{sat}(T_\infty) + P_{sat}(T_\infty) X_\infty} \right) \quad (13)$$

$$X_\infty = 0.622 \left(\frac{RH_{amb} P_{sat}(T_{amb})}{P_{atm} - RH_{amb} P_{sat}(T_{amb})} \right): \text{water content far from the surface (kg}_{\text{water}}\text{kg}^{-1}\text{solid)} \quad (14)$$

$$P_s = aw P_{sat}(T_s) \quad (15)$$

The fruit water activity at the interface (aw) as a function of the moisture contents ($X_s = \text{CPM}/\rho_{ds}$) is reported in table 1.

Heat flux

$$n \cdot (-k \nabla T) = q = h_c (T_\infty - T) - N [C_{pv} (T - T_{ref}) + L_v] PM_{H_2O} \quad (16)$$

where q (W m⁻²) is the heat flux, T_∞ (K) is the drying cabinet temperature, C_{pv} (1000 J Kg⁻¹K⁻¹) is the specific heat of water vapour, L_v is the water latent heat (2256 kJ kg⁻¹) and T_{ref} is the reference temperature equal to 273.15 K. Considering the low air speed inside the dryer, only the natural convection was taken into account. The convective heat transfer coefficient h_c (W m⁻²K⁻¹) depending on the product geometry and the ambient flow conditions, was obtained from the Nusselt number (Nu) by the following equation:

$$h_c = \frac{Nu k_a}{L} \quad (17)$$

being:

$$Nu_t = 0.27Ra^{1/4} \text{ for the top surface} \quad (18)$$

$$Nu_b = 0.54Ra^{1/4} \text{ for the bottom surface} \quad (19)$$

$$Nu_w = \frac{0.67Ra^{1/4}}{[1+(0.492/Pr)^{9/16}]^{4/9}} \text{ for the wall sides} \quad (20)$$

$$Ra = GrPr: \text{ Rayleigh number} \quad (21)$$

$$Gr = \frac{g\beta L^3 \rho_a^2 (T_s - T_\infty)}{\mu_a^2}: \text{ Grashof number} \quad (22)$$

$$Pr = \frac{c_{pa}\mu_a}{k_a}: \text{ Prandtl number} \quad (23)$$

The mesh was refined up to a level for which the calculus improvements were not significant and it was characterized by 1406 triangles and 6 layers of quadrilateral boundary elements characterized by a stretching factor of 1.2 (increase in thickness between two consecutive boundary layers) has been applied to all the boundaries. The Arbitrary Lagrangian-Eulerian (ALE) approach was implemented in the model in order to account both radial and axial shrinkages that occurred during drying. The motion of the deformed mesh was modelled using Laplace smoothing. The shrinkage was incorporated in the model as a function of the moisture content (experimental relationship).

Table 1. Material properties implemented in the model.

Properties	Apple	Banana	Air
Thermal conductivity, k (Wm ⁻¹ K ⁻¹)	0.490-0.443exp(-0.206X) (Lonzano et al., 1979)	0.0065X _w +0.126 (Bart-Plange et al., 2012)	-2.28E-3+1.155E-4T-7.9E-8T ² +4.12E-11T ³ -7.44E-15T ⁴
Density, ρ (kgm ⁻³)	0.636+0.102ln(X) (Lonzano et al., 1979)	980 (Khan et al., 2017)	346.52/T
Specific heat, C_p (Jkg ⁻¹ K ⁻¹)	1000(1.4+3.22X) (Bialobrzewski, 2006)	0.811X _w ² -4.72X _w +1742 (Bart-Plange et al., 2012)	1.05E+3-3.73E-1T+9.45E-4T ² -6.02E-7T ³ +1.28E-10T ⁴
Diffusion coefficient, D (m ² s ⁻¹)	-	-	-2.775E-6+4.479E-8T+1.656E-10T ²
Dynamic viscosity, μ_a (Pas)	-	-	-75.20E-10+4.427E-8T-7.887E-12T ²
Water activity, aw	X=0.098(aw/(1-aw)) ^{0.65} (Iguedjtal et al., 2007)	1/aw=[16.63-0.1212(T+273)/X] ^{0.902} +1 (Phoungchandang et al., 2000)	-
Initial moisture content, (kg kg ⁻¹ _{solid})	5.66	3.55	-

Note: T: temperature (K), X= dry basis moisture content, X_w: wet basis moisture content.

Moisture diffusivity optimization

The distance between numerical and experimental data was considered as an objective function (OF), and defined as following:

$$OF(D) = \int [\bar{C}_{exp}(t) - \bar{C}_{num}(t, D)]^2 dt \approx \sum_i [\bar{C}_{exp}(t_i) - \bar{C}_{num}(t, D)]^2 \quad (24)$$

where t_i represents the drying time steps used in the experimental tests.

It is accepted that the diffusion coefficient can be described by an exponential form considering the temperature moisture dependence and the influence, so the temperature and moisture content dependency of the diffusivity was implemented in the model (Balzarini et al., 2018):

$$D(T, C) = A \times \exp\left(-\frac{B}{T}\right) \times \exp(E \times C) \quad (25)$$

The optimal parameter D value is the one which corresponds to the minimum of OF (least-squares errors criteria) and it was calculated by using the optimization module of Comsol Multiphysics. The calculation procedure was the same described by (Fabbri et al., 2014). Levenberg–Marquardt method with the numerical evaluation of second gradient was

chosen as the optimization algorithm. The parameter values proposed by Balzanini et al., (2018) for the drying of the red chicory, were used as initial trial.

2.2 Analytical method.

Fruit samples were considered as an infinite slab because the thickness (l) of the slice (3 mm) was much less than its initial diameter (30 mm). The moisture diffusivity for an infinite slab was therefore calculated by the well know 2nd Fick's law:

$$\frac{\partial X}{\partial t} = D \frac{\partial^2 X}{\partial x^2} \quad (26)$$

Considering the experimental condition, the thermal gradient effect on the moisture was ignored, the moisture transfer was defined one-dimensional (mass transfer considered symmetric with respect to the center) and the initial moisture content (X_0) was considered uniform. In this way, the analytical solutions, for the average moisture content, with constant diffusivity is given by (Crank, 1975):

$$\frac{\bar{X} - X_\infty}{X_0 - X_\infty} = \frac{8}{\pi^2} \sum_{n=1}^{\infty} \left[\frac{1}{(2n-1)^2} \exp\left(-\frac{(2n-1)^2 \pi^2 D t}{4 \frac{l^2}{z}}\right) \right] \quad (27)$$

Following a common procedure, when the only one term of the series solution is considered (for long drying time), the equation is linearized and D can be determined from the slope of $[\ln[(\bar{X} - X_\infty)/(X_0 - X_\infty)]]$ against time curve:

$$D = 4 \text{slope} \frac{l^2}{\pi^2} \quad (28)$$

RESULTS AND DISCUSSION

The inverse method results were obtained by using a PC equipped with 24 CPU (Xeon5675 64 bit 3.07 GHz) and 24 GB RAM, the calculation time was about 10 minutes.

The apple moisture diffusivities and the temperature calculated (fruit center) over the drying time are reported in figure, for three different drying temperature. Comparable behaviour was observed also for the banana samples. For both fruits, the diffusion coefficient depends on sample moisture and temperature and the estimated equation (25) parameters were $A=0.01$, $B=6002$ and $E=2.1$ and $A=0.03$, $B=7203$ and $E=3.5$ for the apple and banana, respectively. Diffusivity decreases during drying process as a consequence of the temporal moisture reduction. After about 180 min (apple) and 250 min (banana), the diffusivity takes a constant values corresponding to the diffusivity for the equilibrium moisture. Similar results were achieved by Balzarini et al., (2018) that reported that D is high at the beginning and then was kept constant at the final period, for the drying of chicory root cubes. With the increment of drying temperature an increment of the diffusion coefficients (at the equilibrium) was observed, as a consequence of higher water mobility induce by higher drying temperature. During the transient temperature rise period, an inverse relation between diffusivity and drying temperatures was observed. This is probably due to the greater effect of the moisture content respect to the temperature one.

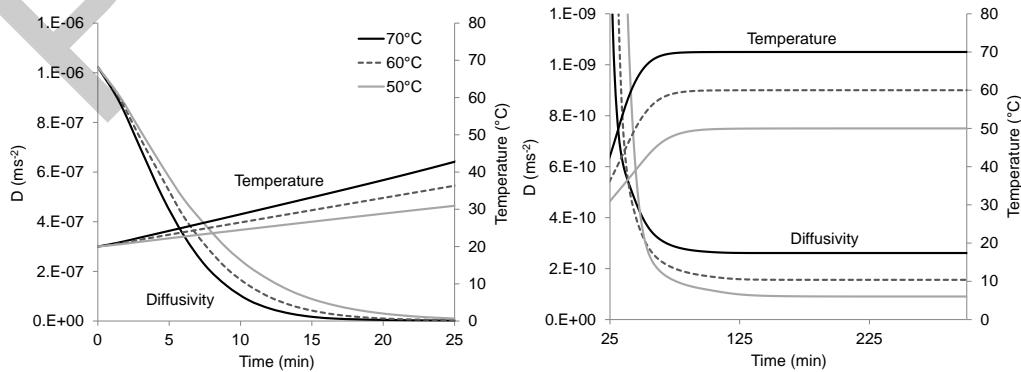


Figure 1. Diffusivity and temperature, calculated at the apple center, over the drying time.

The spatial moisture distribution over the time obtained from simulation results of the apple drying at 70°C is shown in Figure 2. It can be observed also the sample shrinkage, in term of radius and high reduction. After 180 min of drying, the moisture content decreases very slowly and it can be considered reached the equilibrium. Similar behaviour was observed also for the banana samples.

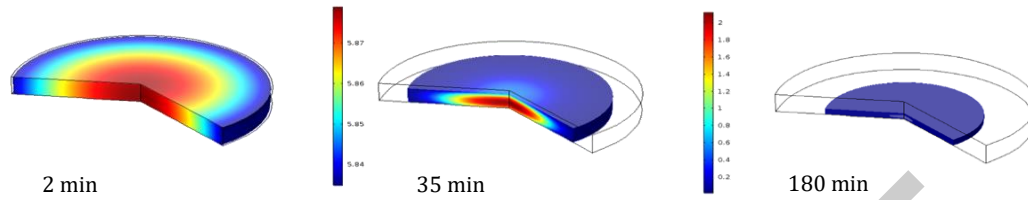


Figure 2. Moisture content over the time calculated for the apple drying at 70°C

The simulated and experimental mean moisture contents over the drying times are shown in figure 3. Simulated data were obtained by using the diffusivity estimated by inverse methods, whilst the experimental ones corresponding to the moisture content values obtained during the experimental phase. The agreement between simulated and experimental values is represented by determination coefficients (R^2) of 0.993, 0.992 and 0.986, and 0.993, 0.993, and 0.976, for apple and banana respectively.

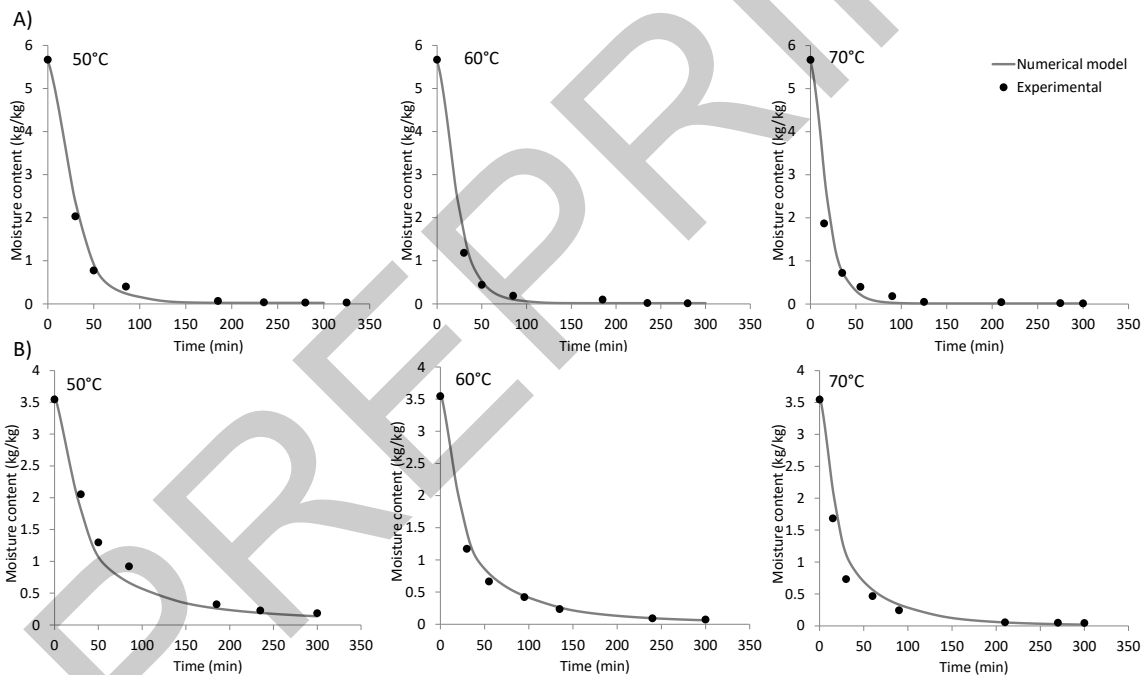


Figure 3. Calculated and experimental mean moisture contents over the drying times: (A: apple; B:banana).

The data obtained by using the inverse methods were compared with those achieved by the classical *slope* method. From the experimental drying data, dimensionless moisture content values were calculated at different drying time and temperature and the *slope* of $\ln[(\bar{X} - X_\infty)/(X_0 - X_\infty)]$ against drying time was taken into account for the calculation of moisture diffusivity (see table 2). The effect of the temperature on the diffusivity was taken into account with an Arrhenius type equation $D = a \exp(-b/T)$. The equation parameters are reported in table 2.

For the data obtained by the inverse method, the calculated mean diffusivity values (in drying time and considering only the thermal equilibrium part) were considered.

Particularly, mean values of $9.72\text{E-}11$ (50°C), $1.59\text{E-}10$ (60°C) and $2.63\text{E-}10$ (70°C) m^2s^{-1} and $5.25\text{E-}11$ (50°C), $7.23\text{E-}11$ (60°C) and $9.98\text{E-}11$ (70°C) m^2s^{-1} , were obtained for the apple and banana, respectively. These values have the same magnitude of those obtained by the slope method and they are in agreement with those reported in literature for similar drying temperature. Particularly, for the banana fruit, Joardder et al., (2014) reported values from $6.63\text{E-}10$ to $1.03\text{E-}10$ m^2s^{-1} (drying temperature of 70°C) while (Demirel and Turhan, 2003) obtained D values ranging $3.8\text{E-}11$ to $1.78\text{E-}10$ m^2s^{-1} for samples dried at temperature from 40 to 80°C . As concern the apple fruit, D values between $6.75\text{E-}10$ and $9.93\text{E-}10$ m^2s^{-1} (drying temperature from 50 to 70°C) were reported by (Beigi, 2016).

Table 2. Diffusivity values obtained by using the *slope* method and parameter of the Arrhenius type equation ($D=a\exp(-b/T)$).

	Apple	Banana
Temperature ($^\circ\text{C}$)	D (m^2s^{-1})	D (m^2s^{-1})
50	$9.36\text{E-}11$	$6.23\text{E-}11$
60	$1.16\text{E-}10$	$7.78\text{E-}11$
70	$1.41\text{E-}10$	$1.06\text{E-}10$
Equation parameters		
a	$3.94\text{E-}10$	$4.16\text{E-}10$
b	-72.43	-97.083
R ²	0.995	0.966

CONCLUSIONS

The inverse method allowed to estimate the effective moisture diffusivity during fruit drying as a simultaneous function of temperature and moisture content. The calculated diffusivity values were then used to solve direct models, in unsteady conditions, showing good agreement with experimental data ($R^2 > 0.97$). The mean values obtained by using the inverse method are in agreement with those obtained by the classical *slope* method, and they are in the same range of those reported in literature. The advantages of proposed inverse technique is that realistic geometries (not simplified) and boundary conditions can be used and that the diffusivity may be introduced as a function of a desirable property (eg. moisture contentment, temperature, porosity). Furthermore the method can be applied to different fruit and vegetable materials during drying.

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