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# Estimation of the mass diffusion and heat/mass transfer coefficients in bay leaves by using the inverse approach

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# Abstract

The thermal and mass transfer parameters are essential to predict the drying kinetics. Unfortunately, these properties are often unavailable in the literature, especially for food products. This work proposes a method for predicting transfer parameters based on the inverse modeling approach. The parameters to be optimized are the external transfer coefficients as well as the moisture diffusivity. The optimization problem consists to minimize the difference between predicted and measured drying kinetics. This study includes modeling and experimental works to predict and measure the drying kinetics. In this study, we are interested in convective drying with hot air and we have used laurel leaves as an example of a product. The predictive model includes coupled phenomena of heat and mass transfers. The experimental tests are carried out in a flow-through drying cell with monitoring of the water loss for different drying conditions (velocity, temperature, and relative humidity).

Thanks to the inverse modeling approach, three parameters are predicted: convective heat and mass transfer coefficients, and the moisture diffusivity. Thereafter, the obtained heat transfer values and moisture diffusivity parameters were implemented on a predictive model to simulate a thin layer of bay leaves for different drying air conditions. The direct model is validated on a set of various thin-layer drying kinetics.

# Keywords

Air drying; inverse modelling; heat and mass transfer; Finite volume method; Parameter estimation;

# Nomenclature

Roman capital letters	
Bi	Biot number

С	Concentration of water vapor in air (kg/m <sup>3</sup> )
CV	Control volume
Ср	Specific heat capacity (J/kg/K)
D	Effective moisture diffusivity (m <sup>2</sup> /s)
Da	Moisture diffusivity coefficient in the boundary layer (m <sup>2</sup> /s)
Μ	Molar mass of water (18g/mol)
N	Number of control volumes
Nu	Nusselt number (-)
L	Width of the leave (m)
Le	Lewis number (-)
ODE	Ordinary differential equation
OF	Objective function (-)
P <sub>v</sub>	Partial pressure of water vapor (Pa)
$P_v^{sat}$	Saturated pressure of water vapor (Pa)
Re	Reynolds number (-)
R <sub>G</sub>	Gas constant (8.314 J/mol/K)
RH	Relative humidity of air (decimal)
S	Surface area of the solid (m <sup>2</sup> )
Т	Temperature (°C)
T <sub>réf</sub>	Reference temperature (°C)
T <sub>0</sub>	Initial temperature (°C)
V	Volume of the solid (m <sup>3</sup> )
Va	Air velocity (m/s)
Х	Moisture content (kg water/kg dm)
Roman small letters	
a <sub>w</sub>	Water activity (-)
dz	Infinitesimal thickness increment (m)
е	Thickness of the product (m)
h	Convection heat transfer coefficient (W/m <sup>2</sup> /K)
i	i-th elementary volume

k <sub>m</sub>	Convection mass transfer coefficient (m/s)			
m	Mass flux density at surface kg/s/m <sup>2</sup>			
n	Number of data points			
p <sub>i</sub>	Constant coefficients of effective moisture diffusivity equation (-)			
t	Drying time (s)			
u	Internal energy (J/m3)			
Z	distance from the center (m)			
Greek letters				
α	Thermal diffusivity (m <sup>2</sup> /s)			
$\Delta H_{v}$	Latent heat of water vaporization (J/kg)			
Δz	Thickness of a compartment/volume (m)			
λ	Thermal conductivity (W/m/K)			
μ	Dynamic viscosity of air (kg/m/s)			
ρ	Density (kg/m <sup>3</sup> )			
Subscripts				
a	Dry air			
cal	calculated			
dm	Dry matter			
е	Eastside face of the CV			
eq	Equilibrium, at end of drying			
est	estimated			
h	Heat transfer			
i	Initial			
m	Mass transfer			
ω	Relative to the drying air			
V	water vapor			
W	Westside face of the CV			

# 1. Introduction

Hot air drying is one of the most common drying techniques used to remove moisture from agroindustrial and food products for preservation purposes and to produce quality material (Mujumdar, 2004). During this process, numerous important modifications take place in the product such as biological, physicochemical, and structural changes that affect the final material quality. The drying mechanism is considered as a process of coupled energy and mass transfer occurring inside the product and at the boundary with drying air, and is affected by numerous parameters, such as the drying air conditions and the properties of the material. To better predict the drying behavior of the product, it is necessary to understand the physical mechanism governing the drying operation. Unfortunately some thermophysical properties are difficult to determine experimentally (e.g. moisture diffusion).

Drying is often considered very energy-consuming. Understanding and predicting thermal and mass transfer phenomena is essential to simulate and improve energy efficiency (Romdhana et al, 2016; Chriyat et al, 2017). Understanding and predicting the phenomena induced on the product to be dried is essential for controlling the product in its form, structure, taste, nutritional quality (Kumar et al, 2012; Aversa et al., 2007). Numerous studies on mathematical drying models available in the literature are divided into two principal categories: empirical and physical models.

On one hand, the empirical models are easiest to use, because the experimental drying data can be solved by regression analysis using a simple fit model such as logarithmic, exponential and quadratic function. Numerous works on empirical models have been published for different agro-industrial and food materials: for Jujube (Chen et al, 2015), rosemary leaves (Mghazli et al, 2017), bay laurel leaves (Ghnimi et al, 2016), apple pomace (Wang et al, 2007), etc. On this topic, various applications for these models have highlighted that they are only specific to the conditions and product under which it is applied (Erbay and Icier, 2010; Onwude et al., 2016c). Therefore, these models are not able to provide a physical description of the drying process because they are not based on the fundamental physical laws governing the mechanism of energy and mass transport inside the product (Ertekin and Firat, 2017; Chou et al, 2000).

On the other hand, the physical models are based on numerical solutions of mass and heat transport equations. The simulation of moisture and temperature distribution is based on scientific knowledge on the physical mechanisms governing the drying phenomena and explains how the operating conditions have effects on product properties. Unlike empirical models, the physical model can be used for a wide range of applications and drying conditions (Kumar et al, 2012; Lambert et al, 2015; Xanthopoulos et al, 2009). Physical models of drying can be developed in different ways. The first approach is a diffusive model for which only moisture content and temperature are determined and the diffusion coefficient is considered global (apparent) because vapor and liquid phases are not separated (Golestani et al, 2013;Kokolj et al.,

2017; Sadeghi et al., 2016; Tank et al., 2014). The second approach is a multiphasic model with liquid and vapor phases; for each phase, a diffusion coefficient is determined, the mass conservation is considered; the gas pressure can be also considered (Dissa et al., 2014; Nicolas et al., 2014; Purlis, 2019).

Models based on global (apparent) diffusion are classified into two types, namely conjugate and nonconjugate problems. Transfers in conjugate models can be governed by two coupled unsteady transfer equations, one for the surrounding fluid and the other for the product being dried (De Bonis et al., 2008; Lamnatou et al., 2009, 2010; Halder et al., 2012). These models have a high accurate quantification that can conduct to enhance the characterization and comprehension of the drying process, but the complexity of the non-linear partial differential equations makes the numerical solution complicated and computational very required. Non-conjugate models can be described by the heat and mass transport inside the material, not taking into account unsteady transport equations in the surrounding air,. In literature, numerous works studied the convective drying process of agricultural materials using non-conjugated physical models for its simplicity and reliability (Ateeque et al, 2014; Lemus-Mondaca et al, 2014; Villa-Corrales et al. 2010, Barati et al, 2011; 2012). In these approaches, knowledge of moisture transfert and heat/mass transfer coefficients at interface is required as input to the model and these cannot still be readily available because of their space, time, moisture and temperature-dependent variables.

Moisture diffusivity is the main variable in governing the diffusion of mass through the product during drying. It describes all possible processes of mass movement inside the material that is a mixture of surface diffusion, liquid diffusion, vapor diffusion, hydrodynamic flow and capillary flow (Kim and Bhowmik, 1995). Knowledge of this parameter is essential in moisture transfer processes modeling. Literature data of the latter parameter are not easily available on account of its variability upon numerous variables, such as moisture content, temperature, shrinkage, porosity and drying conditions. Hence, the analysis of the moisture transfer mechanism is dependent on the assumptions which are taken into account for the determination of the effective moisture diffusivity. According to literature data results, effective moisture diffusivity can be expressed as a function of one or several variables such as drying conditions and product properties (Zogzas et al, 1994). Numerous studies have investigated the impact of material temperature and moisture content on the effective moisture diffusivity coefficient. Silva et al, (2011) studied effective moisture diffusivity that was either constant or variable, in banana drying. They proved that the variability of the moisture diffusivity as a function of sample moisture content is more precise in predicting the drying kinetic than the constant moisture diffusivity. Most authors (Batista et al, 2007; Touil et al, 2014; Karim and

Hawlader, 2005) investigated that moisture diffusivity is expressed in term of moisture content, whereas others (Silva et al, 2013) found it as product temperature-dependent. Variation of the diffusivity coefficient depends on the product temperature and moisture content during drying. Silva et al, 2014; Chandra and Talukdar, (2010) proposed that the variation of the diffusivity coefficient depends on the product temperature and moisture content (D=f (T, X)) during drying. This latter assumption was confirmed by a comparison of drying kinetics with that predicted. Kumar et al. (2015) studied the impact of effective diffusivity as a function of moisture content (MDED), effective diffusivity as a function of product temperature (TDED), and their average on the drying kinetics of banana. They found that the first approach was more suitable to the final stage of drying and the second approach was more suitable to the initial stage of drying. By comparing the simulation results with the experimental results, they concluded that the effective diffusivity depending on moisture content (MDED) is more precise for predicting kinetics in food drying than the effective diffusivity depending on product temperature (Temperature (Temperature (Temperature (Temperature Content (MDED)).

Certain methods in the literature that estimate moisture diffusivity are based essentially on Fick's laws. Besides, the main methods to estimate moisture diffusivity (Zogzas et al., 1994) are explicit solutions to the mass transfer equation, analytical solutions, and numerical solutions.

On one hand, the analytical solution has been extensively used in food for estimation of effective diffusivity for various geometries, however under hypothesis like uniform initial moisture content, negligible shrinkage and external resistance, constant diffusivity and isothermal drying (Chen and Putranto, 2013).

On the other hand, the numerical solution has the potential to evaluate the moisture diffusivity. Generally, the moisture diffusivity as a function of product temperature or/and moisture content or other sample properties being integrated into the numerical model. The moisture diffusivity function is determined by fitting the simulation data to the experimental data, and by selecting values for function parameters that make a better fit. To find desired parameters, it is necessary to define an objective function (OF) which is the difference between computed and observed values. Fabbri et al. (2014) proposed an identification based on finite element models to identify the moisture diffusivity during the hydration process in various food products such as biscuit, salami and flatbread. To estimate the diffusivity of chicory root cubes, Balzarini et al. (2018) described the heat and mass transport during drying with identification based on a finite element

model. The moisture diffusivity was introduced in the model as a function of product temperature and moisture content. Lambert et al (2015) identified the effective moisture diffusivity for pellets on deep-layer drying kinetics, and validated it over a set of thin-layer drying kinetics. They found out that a mismatch between experimental and simulation data at the beginning of the kinetics can be explained from a wrong convective heat transfer coefficient. So, to correct this mismatch, it was essential to estimate the heat and mass transfer coefficient.

The models in this study are non-conjugate heat and mass transfer models. To resolve the heat equation and the equation of mass diffusion at the interface between air and product, convective transfer coefficients (CTC) were introduced as additional parameters at the boundary condition (air-product interface). So, the heat/mass transfer coefficients are required as input variables to the computation and these cannot be readily determined. In the literature, various approaches have been considered for the determination of the CTC such as: (i) Constant values or empirical correlations for simplified flow configurations such as flow around plates, and cylinders (Oztop et al.2008; Aversa et al. 2007), (ii) time-dependent coefficients based on a zero-dimensional heat and mass balance (Lemus- Mondaca et al. 2013; Villa-Corrales et al. 2010), (iii) optimization and regression analysis of experimental and numerical data, (Maroulis et al., 1995; Da Silva et al., 2014) and (iv) flow field simulations using computational fluid dynamics (CFD) (Kaya et al.2008; Esfahani et al. 2014). So far, no publications studies are identifying the heat/mass transfer coefficients and the effective moisture diffusivity as a function of product temperature and moisture content for bay leaves according to experiments. It was therefore considered to be worthwhile to estimate both thermophysical properties at the same time.

The main goal of this study is therefore to determine heat/mass transfer coefficients and the effective moisture diffusivity for predicting the drying kinetics for bay leaves by using a finite element model. This work includes four steps: (i) Experimental determination at the lab-scale of bay leaf properties and drying kinetics at constant conditions, (ii) Development of a non-conjugate model describing the heat and mass transfer mechanisms during drying of bay leaves with effective moisture diffusivity depending on temperature and moisture content and with evaporation rate as an additional term at the air product interface, (iii) Estimate the heat/mass transfer coefficients and effective moisture diffusivity for the product, reducing the differences between simulation results and experimental data and (iv) Comparison of the numerical results of mean moisture content versus time with the observation data over all thin-layer drying kinetics for

validation purposes, to understand the influence of the moisture diffusion and the external heat and mass transfer on the drying process.

# 2. Material and Methods

# 2.1 Sample preparation and drying experiments

Fresh bay leaves used in the drying experiments were collected during the autumn months, from the same tree in a local protected botanical (Mornag-Tunisia) and brought immediately to the laboratory. The samples were washed and kept in a refrigerator at 4°C until the drying trials. The drying process was performed at the laboratory scale using a vertical drying tunnel developed by the Laboratory for Energy, Heat and Mass Transfer (LETTM) at the Tunis Faculty of Sciences (Tunisia). This dryer includes a programmable control system to adjust drying air parameters at given setpoints. The overall layout of the drying tunnel is shown in Figure 1 (Hassini et al, 2007; Azzouz et al, 2016). Once the air drier was at a stable state, the product was placed in the drying chamber. For each experiment, a mass of  $50.0 \pm 0.1$  g of fresh leaves was distributed uniformly over a stainless steel grid in a thin layer of about 3 cm. The grid loaded with the leaves was suspended on a digital balance. The convective drying experiment was based on a three factor face-centered central composite design (FCCCD) consisting of drying temperature (40-70°C), relative humidity (15-35%) and air velocity (1-3 m/s), with three levels for each parameter, as summarized in Table 1. These ranges were chosen because it is known that they are large enough to contain conditions without an excessive effect on product quality (Yagcioglu et al., 1999; Demir et al., 2004; Skrubis, 1982). The mass of the product was weighed during the experiment using a digital balance with a range of 5 kg and accurate to 1 g, placed outside the drying chamber and recorded using a data logger. Drying experiments were pursued until the moisture content of the sample reached the equilibrium state, which was observed when two or three consecutive weighings did not indicate any significant variations or changes in value. Temperature, relative humidity and air velocity inside the pilot drying system were adjusted and controlled automatically. The measurement sensors and data recording and control system were coupled to a computer.

#### 2.2 Model for heat and mass transfer

A physical model has been developed for a thin-layer of bay leaves during convective drying. This model is based on mass and energy conservation. To simplify the model, a number of assumptions are considered:

- > The initial moisture content and temperature inside the laurel leaves samples are uniform.
- > The water evaporation takes place only at the interface of the product.

- > Thermophysical properties of the product are uniform and isotropic
- > Shrinkage and chemical reactions are negligible.
- > The mass and heat transfer are assumed unidirectional (through the thickness of the leave)

A first estimate of the thermal Biot number suggests that the resistance to thermal transfer in the bay leaf is negligible compared to the resistance in the boundary layer. Indeed the thermal Biot (Eq.1) is in a range of about 0.02 to 0.5 if we consider a thickness of ~2 mm, a thermal conductivity between 0.2 and 0.5 W/m/K (Jayalakshmy and Philip, 2010) and a heat transfer coefficient between 5 and 50 W/m<sup>2</sup>/K. To simplify the problem, it make sense to consider only the resistance to heat transfer in the boundary layer. This rapid dimensional analysis is difficult to apply for the mass transfer (Eq.2) since the internal diffusion coefficient (diffusivity) and external one (convective mass transfer coefficient) are unknown. So no simplification will be considered for mass transfer. The resistance to external and internal transfers are taken into account in the model.

$$Bi_h = \frac{h e}{\lambda}$$

$$Bi_m = \frac{\kappa_m e}{D}$$

(2)

## 2.2.1 Mass conservation equation:

The variation of local moisture content is obtained by combining Fick's law with the microscopic mass balance in an infinitesimal control volume:

$$\frac{\partial X}{\partial t} = \nabla \left( D(X,T) \nabla X \right)$$

(3)

Assuming Cartesian symmetry, the differential equation for moisture transfer in Cartesian coordinate system becomes:

$$\frac{\partial X}{\partial t} = \frac{\partial}{\partial z} (D(X,T) \frac{\partial X}{\partial z})$$
(4)

#### 2.2.2 Energy conservation equation

By neglecting resistance to internal heat transfer, the variation of internal energy is due to the supply of external energy by convection and by the moisture release in the form of water vapor.

$$V \cdot \frac{\partial u}{\partial t} = S \cdot h \left( T_{\infty} - T \right) - \dot{m} \cdot S \cdot \left( \Delta H_v - C p_v T \right)$$

(5)

By neglecting the shrinkage, the sensible internal energy u can be expressed as:

$$u = \rho C p \left( T - T_{ref} \right) = \rho_{dm} \left( C p_{dm} + X C p_w \right) \left( T - T_{ref} \right)$$
(6)

# 2.2.3 Boundary and initial conditions:

At the beginning of the process (t=0), the moisture content and the product temperature are uniform:  $X=X_0$ and  $T=T_0$ .

At open boundary (i.e interface between the product surface and the drying aid):

$$\left(\frac{\partial X}{\partial z}\right)_{z=e/2} = \dot{m}$$
 (7)

At the center of the product:

$$\left(\frac{\partial X}{\partial z}\right)_{z=0} = 0 \tag{8}$$

The evaporation rate is expressed as function of the concentration of water vapor difference between the product surface and the drying air, and the mass transfer coefficient  $k_m$  (Mihoubi and Bellagi, 2009; Romdhana et al., 2015).

$$\dot{m} = k_m (C_{z=e/2} - C_{z=\infty})$$

By assuming ideal gas behavior for the gaseous phase, the concentration of water vapor close to the product surface and the concentration of water vapor within the drying medium are expressed respectively by Eqs. (10-11):

$$C_{z=e/2} = \frac{M P_{v,z=e/2} \frac{P_{v,z=e/2}^{sat}}{R_G P_{v,z=e/2}^{sat} T_{z=e/2}} = a_w \frac{P_{v,z=e/2}^{sat}}{T_{z=e/2}}$$

(10)

$$C_{z=\infty} = \frac{M P_{v,\infty} P_{v,\infty}^{sat}}{R_G P_{v,\infty}^{sat} T_{\infty}} = RH \frac{P_{v,\infty}^{sat}}{T_{\infty}}$$

(11)

Finally, the evaporation rate can be expressed as:

$$\dot{m} = k_m \frac{M}{R_G} \left( a_w \frac{P_{v,z=\frac{e}{2}}^{sat}}{T_{z=\frac{e}{2}}} - RH \frac{P_{v,\infty}^{sat}}{T_{\infty}} \right)$$

(12)

#### 2.3 Sorption isotherm measurement

Attention cette section ne présente pas une procédure de mesure, donc l'intitulé ne convient pas **#** Sorption isotherm measurement **#** 

Tu dois absolument décrire ces mesures (avec la méthode des sels saturés) dans la section matériel et méthode Il est préférable d'insérer un tableau avec les données brutes Et ensuite tu peux parler du modèle empirique choisi

Desorption isotherms of bay leaves were performed using the gravimetric static method at three levels of temperature (50°C, 60°C and 70°C) and desorption cycle of water activity  $a_w$  was ranged from 0.5 to 0.88. Equilibrium moisture content (X<sub>e</sub>), temperature and water activity ( $a_w$ ) data were fitted according to four equations: Modified Henderson (Shigehisa, 2015), Karlsson Soininen (Karlsson and Soininnen, 1982), Chung and Pfost (Chung and Pfost, 1967), Henderson (Henderson, 1952). The Karlsson Soininen equation was the best fitting of desorption isotherms with higher coefficient correlation  $R^2=0$ , 98.

$$a_{w} = \exp\left(T.\exp\left(-8.87.X_{e} - 11.79.\sqrt{X_{e}} + 0.37\right) - \exp\left(-0.21.X_{e} - 1.49.\sqrt{X_{e}} + 5.12\right)\right)$$
(13)

Where  $X_e$  is the equilibrium moisture content (kg water/kg.dm),  $a_w$  is the water activity (-), and T is the product temperature (°C).

#### 2.4 Estimation of effective moisture diffusivity and heat and mass Transfer Coefficient

In heat and mass transfer simulation, it is necessary to supply all thermophysical properties and parameters of drying air and product. Some product characteristics and properties are determined experimentally or from value published in literature such as (thickness, density, water activity, water content, etc.). Other physical properties are not readily available (effective moisture diffusivity, external coefficient of heat and mass transfer coefficients) because they depend on numerous parameters such as drying air conditions, water activity, water content and temperature product. The effective moisture diffusivity and the heat transfer coefficient of the sample are estimated by the reverse method by fitting a set of experimental kinetics data obtained in a thin layer and under various conditions of drying air: temperature is between 40 to 70°C, air relative humidity is between 15 to 35 % and air velocity is between 1 to 3 m/s. In this work, the equation of effective moisture diffusivity is a function of the product temperature andof the moisture content (Zogzas et al., 1996; Romdhana et al., 2015):

$$D(X,T_p) = p_0.\exp(-p_1.X + p_2.X.T_p)\exp(\frac{-p_3}{T_p})$$

(14)

Where  $p_0, p_1, p_2$  and  $p_3$  are constants.

The mass and heat transfer coefficients were related by the Chilton- Colburn analogy (Chilton & Colburn, 1934; Cengel and Ghajar 2011; Pasban et al., 2017). Thus, the mass transfer coefficient can be expressed by:

$$k_m = \frac{h}{\rho_a C_{pa} (Le)^{2/3}}$$

(15) Where Lewis number (Le) was defined as:

$$Le = \frac{\alpha_a}{D_a}$$

(16)

# 2.5 Heat transfer Calculation:

The empirical model of convective heat transfer coefficient in the surface of a product is based on a correlation Nusselt that is typically expressed as a function of Reynolds and Prandtl numbers in the laminar flow over sample plates (Karim and Hawlader., 2005; Golestani et al., 2013), were given in Eq.17:  $Nu = 0.664 Re^{0.5} Pr^{0.33}$ 

# (17)

The Nusselt (Nu), Reynolds (Re) and Prandtl (Pr) numbers were expressed as:

$$Nu = \frac{h_h L}{\lambda_a}$$

$$Re = \frac{V_a \, L \, \rho_a}{\mu_a}$$

(19)

$$Pr = \frac{\mu_a C_{pa}}{\lambda_a}$$

(20)

# 2.6 Finite volume method:

To solve the previous partial differential equations (PDE) describing the heat and mass transfer in the drying process of a flat particle, it is useful to mesh the continuous computation area. Finite differences, finite elements and finite volumes are widely used as discretization methods. In this work, the finite volume method (FVM) is used to solve numerically the problem of heat and mass transfer. This approach is useful

for complex geometries (Webley and He, 2000). The 1D continuous domain is split into N finite control volumes  $CV_i$  with N-2 compartments of constant mesh sizes  $\Delta z$  and two boundary compartments (center and boundary layer) of mesh sizes  $\Delta z/2$ . The Eqs. 4 and 5 are integrated over each cell of the mesh. Each control volume has an east and a west (subscripts e and w) interfaces and a control volume center, are shown in

# Figure 2.

The finite interval  $\Delta z$  is defined as:

$$\Delta z = \frac{e}{2(N-1)}$$

(21)

For i between 1 and N, each CV<sub>i</sub> is identified by its representative size defined as:

$$z_{ci} = (i-1)\Delta z \tag{22}$$

The interfaces of control volume (CV<sub>i</sub>) are then:

$$z_{i,w} = z_{ci} - \frac{\Delta z}{2}$$

$$z_{i,e} = z_{ci} + \frac{\Delta z}{2}$$
(23)

The integral form of the mass conservation equation is discretized into N volume integrals in the physical space to obtain the following expressions:

$$\int_{-\dot{u}_{i,w}}^{z_{i,e}} \frac{\partial X}{\partial t} S \, dz = \int_{-\dot{u}_{i,w}}^{z_{i,e}} \frac{\partial}{\partial z} (D \frac{\partial X}{\partial z}) S \, dz$$

(25)

Il n'y a pas de dérivée spaciale de l'équation 5 donc ne nécessite pas une discrétisation par la méthode des volumes finis

$$\int O$$

To simplify the problem, the moisture content is assumed uniform in a control volume  $(CV_i)$  and the internal energy is considered uniform in the global continuous domain, Eqs.25 and 26 become:

$$\frac{\partial X_{i}}{\partial t} = \frac{1}{Z_{i,e} - Z_{i,w}} \left[ D_{i} \frac{\partial X_{i}}{\partial z} \right]_{v}^{t}$$

(27)

[()]A linear approximation of the mass flux is applied to determine the moisture content gradient at the interfaces of each  $CV_i$ , as follows:

$$\frac{\partial X_i}{\partial z} = \frac{X_{i+1} - X_i}{\sum_{i=1}^{n} \frac{\partial X_i}{\partial z}}$$

(29)

$$\frac{\partial X_i}{\partial z} \bigg| , = \frac{X_i - X_{i-1}}{\Delta z}$$

(30)

Then, the moisture diffusivity at west and east interfaces are calculated by using a geometric mean between nodal points:

$$D_{i,w} = (D_{i-1}D_i)^{1/2}$$

(31)

 $D_{i,e} = (D_{i+1}D_i)^{1/2}$ 

(32)

A system of N+1 first-order differential equations (ODEs) is obtained by substituting Eqs. 29 and 30 into Eq. 27 and using the initial and boundary conditions, as:

#### For i=1:

$$\frac{\partial X_1}{\partial t} = \frac{1}{\Delta z^2} D_{1,e} (X_2 - X_1)$$
(33)

For i=2.....N-1:

$$\frac{\partial X_i}{\partial t} = \frac{1}{z_{i,e} - z_{i,w}} \left( D_{i,e} \frac{X_{i+1} - X_i}{\Delta z} - D_{i,w} \frac{X_i - X_{i-1}}{\Delta z} \right)$$

For i=N

Cette equation est fausse!!!! À vérifier

$$\frac{\partial u}{\partial u} = \frac{1}{\left(\frac{e}{2}\right)} \left( h(T_{\infty} - T) - m(\Delta H_v + Cp_v T) \right)$$

J'ai barré l'indice N de la temperature, car il n'a pas de sens puisque la temperature est uniforme, et moi j'ai mis Tp avec p comme product.

#### 2.6 Numerical and optimization strategies:

These sets of ordinary differential equations (stiff systems) were solved in MATLAB® software at each node using ode15s solver that is a method based on backward differentiation formulas for stiff problems (Matlab program). At each step, the local error in the ith component of the solution was determined with the relative and absolute error tolerances according to Table 2. Therefore, the subroutine "fminsearch" was used to estimate the moisture diffusivity parameters and the heat transfer coefficient. This latter function applies the Nelder–Mead simplex method to minimize the objective function (OF) value. The OF Eq. (34) is defined as the sum of the square of the errors between the simulated and experimental moisture content data. These parameters, once estimated, were employed to solve the model equations and determine moisture content fields in the drying particle.

OF = 
$$\sum_{i=1}^{n} (X_{exp} - X_{sim})^2$$
 (34)

Where n is the number of data points,  $X_{exp}$  is the experimental moisture content (kg water/kg.dm) and  $X_{sim}$  is the simulated moisture content (kg water/kg.dm).

#### 3. Results and discussion

# 3.1 Estimation of moisture diffusivity coefficient

## On dit qu'on calcule h à partir de la correlation 17

#### et on estime D par la methode inverse

#### je pense que l'estimation de h fragilise ce travail (car le tab 3 est un peu troublant)

The heat transfer coefficient by convection is estimated from correlation 17. The obtained values vary between 15, ~21 and 27 W/m2/K, respectively for air velocity values of 1, 2 and 3 m/s. For each velocity value the number of Pr varies very slightly ( $0.7 \sim 0.9$ ). However, there is a net variation in the transfer coefficient as a function of the air flow around the sheet to be dried.



Figure? Calculation of heat transfer coefficient by convection (Pr=0.73 to 0.97)

These findings are in the range of those reported in literature for various products and drying conditions by optimization such as acai berry waste (fruit stone) from 47.78 W/m<sup>2</sup>/C at 42°C to 42.55 to 57 W/m<sup>2</sup>/C (Nagata et al. 2020), slice of potato, apple and carrot from 25 to 90 W/m<sup>2</sup>/C for air velocity range of 1–5 m/s and an air temperature range of 40-60°C under forced convection (Ratti and Crapiste. 1995), products with cylindrical shape from 15.2 W/m<sup>2</sup>/K at 47.9°C to 28.3 at 66.9°C (Silva et al. 2014) and others. Inspection of Table 3 indicates that the values of h<sub>est</sub> are affected by the various drying air conditions. It can be noticed that the values of h<sub>est</sub> increases with the rise in the air velocity and in the air temperature, since the higher in air velocity and air temperature raises the drying potential for the mass transfer and hence the drying rate. In contrast, the heat transfer coefficient diminishes with the rise in air relative humidity, because the greater in the latter parameter decelerate the drying capacity for the transport of moisture. The calculated heat transfer coefficient (h<sub>cal</sub>) based on Nusselt correlation values ranged between 14.2 and 25.6 W/m<sup>2</sup>/K under various conditions (Table 3). It is seen that the h<sub>cal</sub> is not sensitive to the variation in air temperature and in air relative humidity and in the physical properties of fluid. Comparing these findings with those estimated according to the inverse method, it is observed a wide variation between the both data. The reasons for this variation may be due to the dependence of h<sub>eat</sub> only of air velocity, physical properties of air

 $(C_{pa}, \rho_a, \lambda_a \text{ and } \mu_a)$  and characteristics of the physical system, and does not take into account the non-regular product geometries, the operating conditions  $(T_a, RH_a)$  and transient characteristics of heat processes (Rahman and Kumar. 2006; Tolaba et al. 1988).

In this case, an exponential form of moisture diffusivity considering the moisture content and temperature dependence were tested. The initial moisture diffusivity parameters were selected on the base of those reported in literature (Zogzas et al., 1996; Romdhana et al, 2016). The identified global optimum values are:

$$D(X,T_P) = p_0 \exp(-p_1 X + p_2 X T_P) \exp\left(\frac{-p_3}{T_P}\right)$$
(33)  

$$p_0 = 4.7496 \times 10^{-7} \mp 0.27 \times 10^{-7}$$

$$p_1 = 14.0918 \mp 0.0082$$

$$p_2 = 0.0482 \mp 0.0001$$

$$p_3 = 2.9133 \times 10^3 \mp 0.0166 \times 10^3$$

Thereafter, the moisture diffusivity parameters were implemented in the model on the base of the equation proposed beforehand. It is noted that the impact of the moisture content (parameter p1) and of the temperature (parameter p3) appears to be very important and the diffusivity tends to decrease with decreasing moisture content over time and with decreasing temperature. Moreover, the pre-exponential factor ( $p_0$ ) seems to be very restricted. Similar results were reported also by (Lambert et al., 2015; He et al., 2019) who declared that the diffusivity follows the temperature and moisture content tend.

The values of moisture diffusivity coefficient calculated by Eq. (33) for bay leaf is ranged from  $5.15 \times 10^{-11}$  to  $1.65 \times 10^{-9}$ m<sup>2</sup>/s. In this case, these values are comparable with the values of biological products between  $10^{-11}$  and  $10^{-9}$  (Saravacos and Maroulis, 2001; Panagiotou et al., 2004), disposable in the literature. Particularly, (Doymaz et al, 2011) and (Ghnimi et al, 2015) have determined moisture diffusivity coefficients by using the analytical solution of Fick's second law for bay leaf. The first author found values ranging from  $9.38 \times 10^{-12}$  to  $2.07 \times 10^{-11}$  m<sup>2</sup>/s in temperature ranging from 50 to  $70^{\circ}$ C and in constant air velocity 2 m/s, whilst for second author reported a variable value ranging from  $1.21 \times 10^{-10}$  to  $5.27 \times 10^{-10}$  m<sup>2</sup>/s in temperature varying from 45 to  $70^{\circ}$ C, in air relative humidity varying from 5 to 45% and at three air velocities 1, 1.5 and 2 m/s. Other authors were used the numerical solution of Fick's second law based on the inverse method to estimate the moisture diffusivity coefficient for others products such as Fabbri et al. (2014) reported a three moisture diffusivity values being  $1.792 \times 10^{-11}$ ,  $6.804 \times 10^{-11}$  and  $3.857 \times 10^{-12}$  m<sup>2</sup>/s for flat bread, biscuits and salami respectively and Cevoli et al. (2020) mentioned a variable value from 2.61  $\times 10^{-10}$  (T=25°C) to  $7.95 \times 10^{-8}$  (T=178°C) m<sup>2</sup>/s for cake baking.

The Biot numbers (Bi<sub>h</sub> and Bi<sub>m</sub>) are dimensionless quantities used to characterize controlling mechanisms in heat and mass transfer calculations. They are the ratio of the external and internal resistances to heat or mass flux. The knowledge of the both numbers during drying is very important to select an appropriate model. Table 3 shows that the estimated heat transfer value is in a range of about 6 to 58.3 W/m<sup>2</sup>/K Bi<sub>m</sub> thermal Biot is less than 1. Several food products have a Bi<sub>h</sub> less than one such as apple (Golestani et al, 2013) and wheat (Giner et al., 2010). The bay leaf is a poorly conductive ( $\lambda_{max} \sim 0.5$ ) and it have a small thickness, so these product characteristics permitted to have a small value of a thermal Biot. In this condition, the thermal resistance at the surface of the product (convection resistance) exceeds the thermal resistance inside of the product (conductive resistance), this indicates that the temperature interior the material does not vary significantly in space (uniform temperature). This fact permitted to confirm the assumption which was used during the developed of the model "the internal conductive resistance can be neglected and it can be assumed that the product is isothermal". In accordance with the analogy between the transfer processes, the mass transfer Biot number (Bi<sub>m</sub>) is an analogous dimensionless quantity to Bi<sub>h</sub> and have the same role for mass transport as the thermal Biot number has for heat transport. Table 3 shows that the Biot mass transfer is in a range of about  $9.42 \times 10^4$  to  $2.69 \times 10^6$ . It is noticed that the Bi<sub>m</sub> is greater than one. This signifies that the "internal resistance to mass transfer" dominates the mass transfer process, and the "external resistance to mass transfer'' can be neglected because it is small relative to the first term (Parti, 1994). The decrease in moisture content from the X<sub>i</sub> to X<sub>eq</sub> increased the mass transfer Biot number. In this study, it is also noticed that the mass transfer Biot number is much greater, however the thermal Biot number is very small. The mass transfer Biot number is six magnitudes greater than heat transfer. The disparities between the both numbers are the consequences of the relatively lower mass diffusivities in solid materials (Zhang, 2013).

#### 3.2 Model validation

The developed physical model was validated by comparing the experimental data and model predictions. Figures 3-7 displays the comparison of the measured values of moisture content with the predicted results. Mentioned figures shows that the present physical model can predict the temporal evolution of the mean moisture content with acceptable accuracy for a set of numerous thin layer drying kinetics under different drying conditions. This model takes into account the dependence on the air temperature, air relative humidity and air velocity, supplied an appropriate equation of effective moisture diffusivity Eq. (33) and estimated values of heat transfer coefficient (Table 3). The statistical indicators presented in Table 3 are excellent to describe the mass and heat transfer. Particularly, the object function value for the trials is 0.8866

with a relative and absolute tolerance of 10<sup>-4</sup>. It is illustrated that the experimental and simulation data are in good agreement. It can be also inferred that the simultaneous heat and mass transfer model taking into account the effective moisture diffusivity as a function of product temperature and moisture content, can adequately be employed to describe the drying kinetics of bay leaves during convective drying into the wide range of drying conditions. The mismatch observed between the simulated and experimental data, can be caused by the miss of taking into account the change of the product geometry and shape during drying and the variations of the food proprieties, in the present model simulations. The analysis of the transfer phenomena given here is based only on the study of the time evolution of average moisture content. Figure 3 indicated that the repetition of drying kinetic for the center drying condition is found good repeatability with a difference not exceeding 6%. The results in Figures 4-5 revealed that a larger quantity of water is removed in the higher air temperatures and velocities. This comportment is due to the reliance of the effective moisture diffusivity on temperature. It can be seen that an increase in differences temperature between air sample and stream corresponds to an increase in the heat flux to the inside of the sample and thus to higher moisture diffusivity. Furthermore, an increase in air velocity is coupled with an increase in heat transfer coefficient (Table 3), producing in turn to a higher temperature and thereby to an increased diffusivity and increased water removal. Figure 6 shows that water removal is decreasing for increasing air relative humidity. Also, higher air relative humidity corresponds to a decrease in heat transfer coefficient (Table 3), accelerating in turn the decrease of temperature and thereby to the decrease of water removal. Finally, it can be inferred that both moisture content and temperature have an important influence on the evolution of diffusivity.

#### 4. Conclusions

In this study, the physical model was developed based on a simultaneous heat and mass transfer to study the drying convective process of a thin layer of bay leaves. The heat transfer coefficient and the effective moisture diffusivity, as a function of moisture content and product temperature, were identified by inverse modeling approach. When these estimated parameters were introduced in the direct numerical model simulating the thin layer of bay leaves, the minimum value of the distance between the simulated and experimental results was 0.8866. Moreover, the simulation results indicated a good agreement with the experimental data for a series of drying kinetics confirming that the physical model can be predicted the heat and mass transport phenomena during the drying. The benefits of the proposed reverse method are that the heat transfer coefficient may be identified and used and that the moisture diffusivity can be implemented as

a function of a desirable product property (temperature, shrinkage, moisture content...). Furthermore, this model is computationally efficient and thus it is for appropriate for optimizing the quality of the drying convective process.

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# **Conflict of interest**

The authors report no conflict of interest. The authors alone are responsible for the content and 333 writing of the paper.

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# Tables:

Run	Ta (°C)	RH (%)	Va (m/s)
1	40	15	1
2	70	15	1
3	40	35	1
4	70	35	1
5	40	15	3
6	70	15	3
7	40	35	3
8	70	35	3
9	40	25	2
10	70	25	2
11	55	15	2
12	55	35	2
13	55	25	1
14	55	25	3
15 <sup>C</sup>	55	25	2
16 <sup>C</sup>	55	25	2
17 <sup>C</sup>	55	25	2

**Table 1.** Convective drying conditions of the experiments (FCCCD)

<sup>C</sup> Center point.

Table 2. The value of different Thermo physical Properties and the statistical indicators

Thickness	2.10 <sup>-3</sup> m
Characteristic length	0.07 m
Density of bay Laurel leaves	830 kg/m <sup>3</sup> (Measured)
Initial temperature	20°C
specific heat capacity of dry matter	3.580 J.kg <sup>-1</sup> .K <sup>-1</sup> (S. S. Chen and M. Spiro)
specific heat capacity of liquid water	4180 J.kg <sup>-1</sup> .K <sup>-1</sup>
Latent heat of vaporization	2.357 10 <sup>6</sup> J. kg <sup>-1</sup>
Coefficient of mass transfer by convection	Chilton- Colburn analogy (Chilton & Colburn, 1934)
Coefficient of heat transfer by convection	Equation 17
Water activity	Karlsson Soininen equation (Karlsson and Soininnen, 1982)
Effective moisture diffusivity	The present work
number of volumes of finite volume	20
Absolute tolerance	10-4
Relative tolerance	10-4
Thermal conductivity of air	CoolProp (C++ library)
Density of air	CoolProp (C++ library)
Specific heat capacity of air	CoolProp (C++ library)
Dynamic viscosity of air	CoolProp (C++ library)
Mass diffusivity of air-vapor	CoolProp (C++ library)
Thermal diffusivity of air	CoolProp (C++ library)
Wet bulb Temperature	Psychrometric Diagram viewer 3.5.0
OF	0.886

Table 3. Heat convective transfer coefficient and the Biot number of mass transfer.

Run	Ta (°C)	RH	Va (m/s)	h <sub>est</sub>	h <sub>cal</sub>	$\frac{1}{m(X_0, T_{wb}) \times 1\ell_m(X_{eq}, T_{wb}) \times 1\ell}$	
		(%)		(w/m²/K)	(w/m²/K)		
				supprimer	supprimer		
				cette	cette		
				colonne	colonne		
1	40	15	1	7.2	14.8	5.6	6.10
2	70	15	1	33.5	14.5	5.91	1.54
3	40	35	1	6	14.7	2.71	3.89
4	70	35	1	14.5	14.2	0.94	4.42
5	40	15	3	8.5	25.6	6.61	7.20
6	70	15	3	58.3	25.1	10.1	26.9
7	40	35	3	6.8	25.5	3.07	4.44
8	70	35	3	19	24.5	1.21	5.88
9	40	25	2	7	20.9	3.97	5.11
10	70	25	2	23.6	20.3	2.33	8.68
11	55	15	2	15.6	20.8	5.63	9.54
12	55	35	2	8	20.5	1.33	2.75
13	55	25	1	7.8	14.6	1.88	3.92
14	55	25	3	12.8	25.3	3.08	6.50
15 <sup>C</sup>	55	25	2	9.3	20.6	2.20	4.54
16 <sup>C</sup>	55	25	2	9.3	20.6	2.25	4.69
17 <sup>C</sup>	55	25	2	9.3	20.6	2.19	4.69

at different drying conditions.



Figure 1. Overall layout of the experimental dryer.



Figure 2. Schematic of finite volume method discretization of calculation domain.





Figure 4. Comparison of temporal evolution of mean moisture content with simulation data at different drying air temperatures.



Figure 5. Comparison of temporal evolution of mean moisture content with simulation data at different drying air velocities.



# Figure 6. Comparison of temporal evolution of mean moisture content with simulation data at different drying air relative humidity.

