# AN INVERSE METHOD FOR DETERMINING THE INSTANTANEOUS DIFFUSION COEFFICIENT OF PUMPKIN CUBES DURING DRYING PROCESS

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***Abstract*— In food and biological materials, most methods used to determine the effective moisture diffusivity do not provide reliable results. In this work, an experimental approach to determine the instantaneous diffusion coefficient of pumpkin cubes during drying process was used. We use the inverse method by combining the Levenberg-Marquardt optimization technique with analytical solutions of Fick's second law of diffusion. A series of drying kinetics experiments are performed followed by the analytical identification of moisture diffusivity. All experimental runs were carried out according to response surface based on central composite design, and the results obtained were validated.**

***Keywords*— drying kinetics; hot air drying; moisture diffusivity; potimarron cube.**

## I. INTRODUCTION

Drying is a unitary operation involving a transfer of material (the water impregnating the product passes to the vapor state and is recovered by the air) and a thermal transfer (a supply of heat allows the phase change of the product liquid). Thin-layer drying of fruits and vegetables is dependent on the product's kind, pre-treatment, and drying processes. An appropriate mathematical model, such as the liquid diffusion model, which contains the diffusion equation, can be used to describe a specific drying process (Bird *et al.*, 2001; Cunningham *et al*., 2007).

For arbitrary geometry and, in particular, variable thermo-physical properties, the equation of Fick's second law of diffusion must be numerically solved (Gastón *et al*., 2002; Carmo and Lima, 2005; Wu *et al.*, 2004). However, the diffusion equation has an analytical solution under particular assumptions (infinite plate, cylindrical and spherical geometries, constant thermo-physical parameters) (Luikov, 1968). Some researchers employ these solutions to describe the thin layer drying of various fruits and vegetables products (Lima *et al*., 2004; Ruiz-López and García Alvarado, 2007; Hacihafizoglu *et al*., 2008).

An appropriate mathematical model should be developed to describe the drying kinetics of biological materials in order to determine thermo-physical characteristics such as effective diffusivity coefficient. The thermophysical properties can be determined using empirical models, which are usually simple regressions (Doymaz, 2017; Demirhan *et al*., 2018; Benseddik *et al*., 2018; Çalışkan, 2020). However, a general optimization strategy based on the inverse method can be utilized in the situation of the liquid diffusion model (Silva *et al*., 2010, Mariani *et al*., 2008; Da Silva *et al*., 2009).

Research on the experimental estimation of effective moisture diffusivity of agri-food products based on the inverse method is active and widespread in the in Argofood processing sector. Mariani *et al*. (2008, 2009) used a numerical solution of the diffusion equation to propose an optimization approach for estimating the apparent thermal diffusivity value of bananas during the drying process. Zheng *et al*. (2016) estimate the effective moisture diffusivity in lumber during drying using genetic algorithms. Lentzoua *et al*. (2019) employed A moving-boundary FEM model as an inverse problem to estimate water diffusivity and peel resistance incorporating shrinkage. Da Silva *et al*. (2009) used an analytical solution of the diffusion equation to determine the effective diffusivity of mushrooms during drying process. The objective of this work is to apply the inverse method for evaluating instantaneous values of mass diffusivity based on the water content of the product. The method aims at determining the instantaneous mass diffusivity during hot air drying of the cubes of the pumpkin.

## II. MATERIALS AND METHODS

### A. Preparation of samples

The raw material for potimarron pumpkin comes from a local market in La Rochelle, France. The pumpkins are kept in a refrigerator at 4 ° C in the laboratory until drying experiments are carried out. Before beginning any experiment, the pumpkin pieces are sliced into cubes using a culinary aid equipment according to the experimental design. The moisture content of fresh pumpkin was determined using a halogen desiccator MB45 and an oven at 105°C for 24 hours (AOAC, 1990). It has a moisture

Figure 1: Pilot dryer

content of 87 % on a wet basis.

### B. Experimental setup

To investigate the impact of drying parameters on moisture diffusivity during hot airflow drying, the experiments are done using an experimental pilot dryer. The Fig. 1 shows the setting up of a pilot dryer: (1) Compressed air inflow; (2) Flow Control Valve; (3) Heater; (4) Distributor of air; (5) Tray for drying; (6) Balance; (7) Interfaces between humans and machines (HMIs); (8) Logic Controller with Programmability. The pilot drier ensures long-term drying with temperature (40-80 °C) and air velocity (2-15 m/s) that are clearly defined and properly controlled. The dryer operates in an open loop and is controlled by a programmable controller. Temperature and air velocity are adjustable parameters. The air flow passes perpendicularly, from top to bottom, through the pieces of the pumpkin placed on a cup of ten centimeters in diameter. To control the variation in the properties of the air in contact with the product, all the drying experiments are carried out on a monolayer of the pieces of the pumpkin.

### C. Experimental design and statistical analysis

To study the effect of three operating factors on our convective drying, we have retained and favored: the drying temperature (between 40 and 80 ° C), the drying air velocity (2 - 15 m / s) and the cube thickness (between 0.2 - 1.4 cm).

After having chosen the operating parameters which are the most influential and due to a large range of possible variation of each parameter, it is advisable to proceed to a statistical study method capable of identifying the operating domain of the parameters at the lowest cost of experimentation. Thus, during the present study, the method adopted is the composite plan centered at five levels; it is a composite plan centered with three factors, each with five different levels. The experimental plan carried out is presented in Table 1. This is to arrive at suitable samples according to the 18-point design of experiments given in Table 2.

The relationship between the independent variables, air temperature drying ($\boldsymbol{x_1}$), air velocity ($\boldsymbol{x_2}$), and cube thickness ($\boldsymbol{x_3}$), is mathematically formulated as a polynomial model, which gives the dehydration ratio ($Y$) as a function of these factors.

A second order polynomial equation is presented in the following form:

$$y = a_0 + \sum_{i=1}^{3} a_i x_i + \sum_{i=1}^{3} a_i x_i^2 + \sum_{i=1}^{3} \sum_{j>i}^{3} a_{ij} x_i x_j \quad (1)$$

Table1. Central composite design with three factors and five levels used for RSM

| Variables | Coded levels | | | | |
|---|---|---|---|---|---|
| | **−1.68** | **-1** | **0** | **1** | **1.68** |
| $\boldsymbol{x_1}$ | 40 | 48.1 | 60 | 71.9 | 80 |
| $\boldsymbol{x_2}$ | 2 | 4.6 | 8.5 | 12.4 | 15 |
| $\boldsymbol{x_3}$ | 0.2 | 0.44 | 0.8 | 1.16 | 1.4 |

$\boldsymbol{x_1}$= Airflow temperature [oC], $\boldsymbol{x_2}$= Airflow velocity [m/s], $\boldsymbol{x_3}$ = Cube thickness [cm].

Table2. Experimental results of central composite design with three factors and five levels

| Run N° | Factors | | | | | | Responses | | |
|---|---|---|---|---|---|---|---|---|---|
| | Coded variables | | | Uncoded variables | | | Effective average diffusion coefficient Y | Starting accessibility $\delta W_s$ | Starting accessibility ratio $\delta W_s$ |
| | $x_1$ | $x_2$ | $x_3$ | $T[°C]$ | $V[ms$ | $L[cm]$ | $[m^2/s]$ | $gH_2o/gMS$ | [%] |
| 01 | 0 | 0 | 0 | 60 | 9 | 0.8 | 12.46 x 10-9 | 0.7653 | 0.1814 |
| 02 | -1 | -1 | -1 | 48.1 | 5 | 0.44 | 37.02 x 10-9 | 0.7605 | 0.1807 |
| 03 | + 1 | -1 | -1 | 71.9 | 5 | 0.44 | 33.10 x 10-9 | 0.7763 | 0.1845 |
| 04 | -1 | + 1 | -1 | 48.1 | 12 | 0.44 | 18.99 x 10-9 | 0.9623 | 0.1815 |
| 05 | + 1 | + 1 | -1 | 71.9 | 12 | 0.44 | 32.55 x 10-9 | 0.9192 | 0.1754 |
| 06 | -1 | -1 | + 1 | 48.1 | 5 | 1.16 | 52.39 x 10-9 | 0.9833 | 0.1877 |
| 07 | + 1 | -1 | + 1 | 71.9 | 5 | 1.16 | 39.14 x 10-9 | 0.8609 | 0.1851 |
| 08 | -1 | + 1 | + 1 | 48.1 | 12 | 1.16 | 61.55 x 10-9 | 1.0365 | 0.1858 |
| 09 | + 1 | + 1 | + 1 | 71.9 | 12 | 1.16 | 44.89 x 10-9 | 1.1544 | 0.1826 |
| 10 | 0 | 0 | 0 | 60 | 9 | 0.8 | 25.26 x 10-9 | 1.1598 | 0.1835 |
| 11 | 0 | 0 | 0 | 60 | 9 | 0.8 | 63.23 x 10-9 | 0.8459 | 0.1797 |
| 12 | -1.68 | 0 | 0 | 40 | 9 | 0.8 | 24.30 x 10-9 | 0.8565 | 0.1820 |
| 13 | +1.68 | 0 | 0 | 80 | 9 | 0.8 | 25.93 x 10-9 | 0.8974 | 0.1841 |
| 14 | 0 | -1.68 | 0 | 60 | 2 | 0.8 | 45.91 x 10-9 | 1.0865 | 0.1877 |
| 15 | 0 | +1.68 | 0 | 60 | 15 | 0.8 | 33.21 x 10-9 | 0.9877 | 0.1863 |
| 16 | 0 | 0 | -1.68 | 60 | 9 | 0.2 | 31.41 x 10-9 | 0.9344 | 0.1832 |
| 17 | 0 | 0 | +1.68 | 60 | 9 | 1.4 | 50.98 x 10-9 | 1.7317 | 0.1795 |
| 18 | 0 | 0 | 0 | 60 | 9 | 0.8 | 30.05 x 10-9 | 1.2087 | 0.1843 |

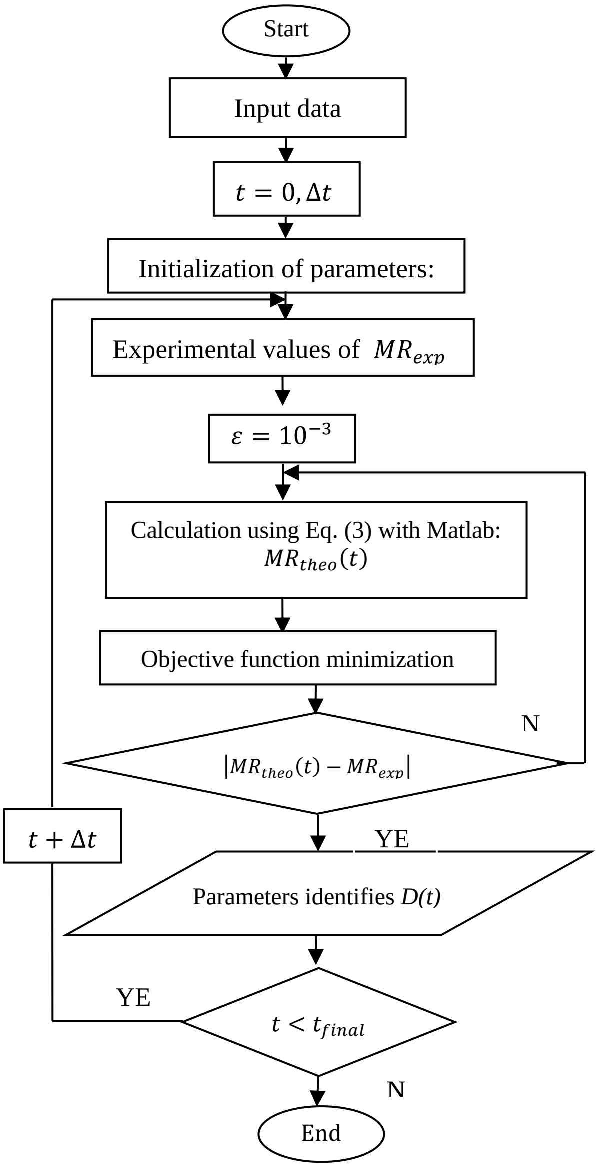


Figure 2: Simplified algorithm for the main program of the inverse method.

**D. Modeling of dehydration kinetics**

Diffusion is considered to be the primary mode of water movement from the solid's center to its surface. Therefore, the rate of mass transfer via pure diffusion is related to the gradient of water concentration, as well as the effective diffusion coefficient.

The determination of this the latter is essential to describe the process of mass transfer by the equation of the second law of Fick is expressed by (Crank, 1975):

$$\frac{\partial MR}{\partial t} = \nabla\left[D_{eff}\ \nabla MR\right] = D_{eff}\ \nabla^2 MR \qquad (2)$$

The cube of potimarron pumpkin can be considered of as a uniform plaque evolving in a non-stationary system with a uniform initial distribution and equal concentrations at the surface. Crank (1975) developed an analytical solution (Eq. 3) for Fick's second law in the condition of an infinite plate with an initial assumed uniform moisture distribution, a constant diffusivity, a negligible shrinkage, and external resistance.

$$MR = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{exp\left(-(2n+1)^2\pi^2 D_{eff} t/4L^2\right)}{(2n+1)^2} \qquad (3)$$

The moisture ratio $(MR)$ can be expressed as follows:

$$MR = \frac{M_t - M_e}{M_0 - M_e} \qquad (4)$$

where $M_0$, $M_e$ and $M_t$ are the initial, the equilibrium moisture content (%, db) and at drying time (t), respectively, $t$ is the drying time (s), $D_{eff}$ is the effective diffusivity (m²/s), $L$ is the half of the thickness of the cube (m) and n is a positive integer.

The experimental data employed in Eq. (3) exclude those concerning the points close to $t = 0$; the model extrapolation so obtained resulted in assessing $W_0$ as generally distinct from the initial humidity content $W_i$. The difference between $W_i$ and $W_0$ at the starting point decoded the superficial water quickly removed from the surface, independent of the diffusion process. The starting accessibility $\delta W_s$ and the starting accessibility ratio $\delta W_s\%$ were then defined by (Mounir and Allaf, 2008), respectively, as (Fig. 4):

$$\delta W_s = W_i - W_0 \qquad (5)$$

and

$$\delta W_s\% = \frac{W_i - W_0}{W_i} \qquad (6)$$

The taking into account of all the analytical solution can be done only by numerical resolutions. This technique makes it possible to develop a protocol for identifying the effective diffusivity by minimizing, using an algorithm, a criterion which is the sum of the squares of the differences between the theoretical reduced water content and that measured.

$$critere = \frac{1}{N}\sum_{i=1}^{P}\left(MR_{théo}(i) - MR_{exp}(i)\right)^2 \qquad (7)$$

where $N$ is the total number of measurements; $MR_{exp}(i)$ is the $ith$ moisture ratio observed experimentally over time $MR_{théo}(i)$ is the ith moisture ratio calculated over time.

The theoretical moisture ratio $MR_{théo}$ is determined from Eq. (3) without any restriction.

**E. Inverse method**

When material constants are difficult to determine physically, the inverse method, also known as a type of curve fitting process, is generally implemented in engineering fields. This method is created and applied in this research to iteratively determine the instantaneous diffusion coefficient on the basis of the experimental drying kinetics curves. It is performed by minimizing at each iteration an objective function representing the difference between the measured outputs of the process and the outputs of the model, based on a nonlinear least squares algorithm. The flowchart is shown in Fig. 2.

The numerical values thus found are compared with the experimental values, taken at the same time, by means of the least squares criterion. A minimization procedure of this criterion is adopted to find the final estimated values.

## III. RESULTS AND DISCUSSION

Typical drying curves $MR_{theo}$ as a function of time are

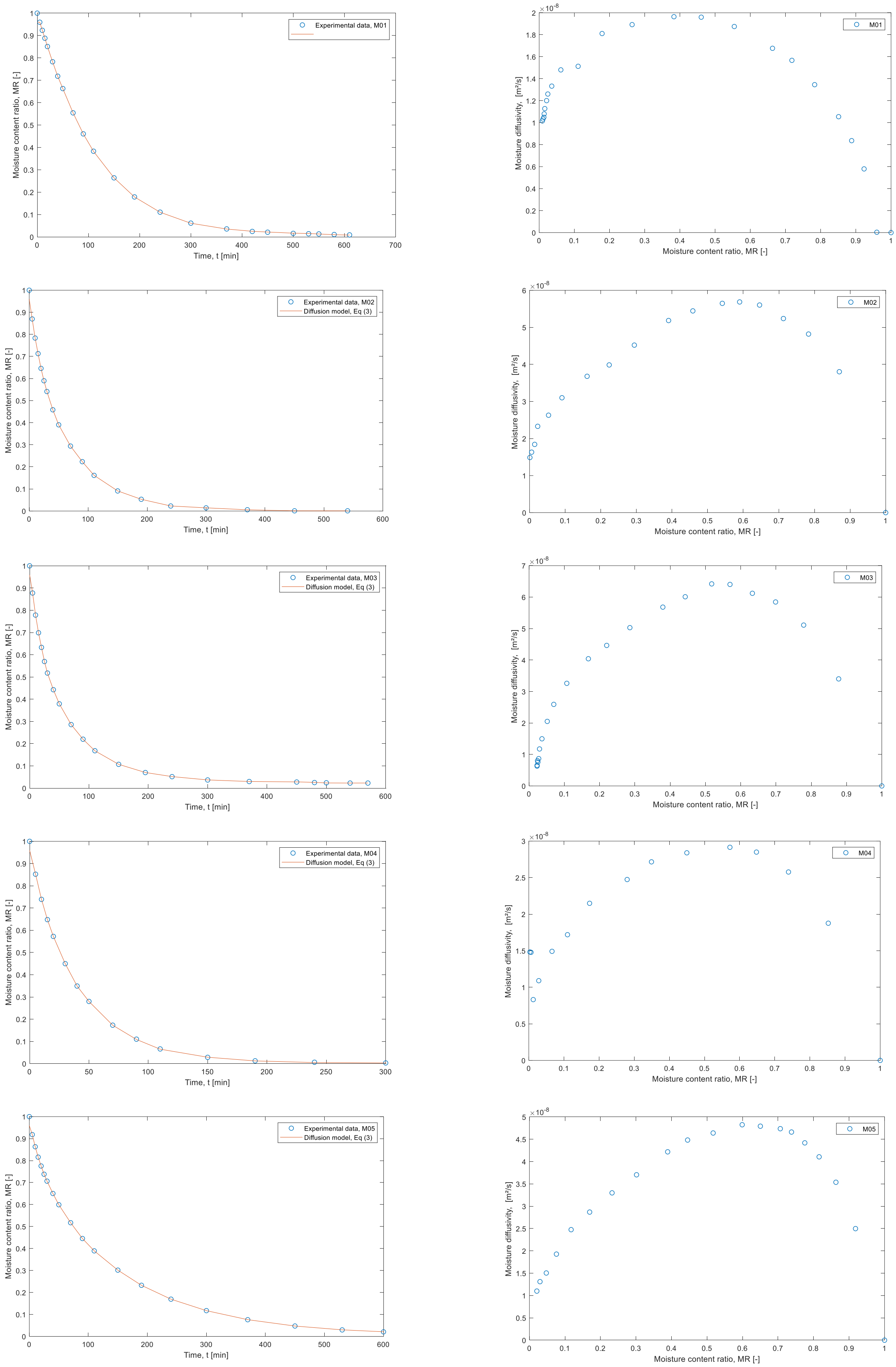


Figure 3: Moisture content ration and instantaneous diffusion coefficient during the drying process of the pumpkin cubes for the ten first manipulations of central composite design.

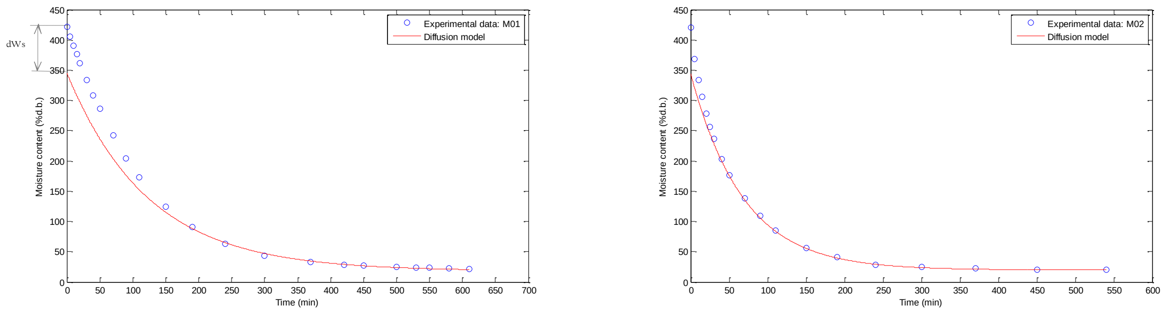


Figure 3: Moisture content ration and instantaneous diffusion coefficient during the drying process of the pumpkin cubes for the ten first manipulations of central composite design.

shown in the figures to the left of Fig. 3. The instantaneous diffusion coefficients calculated by the inverse method are shown in the figures to the right of Fig. 3. The agreement perfect between the measured moisture content ratio $MR_{exp}$ expand that calculated $MR_{theo}$ by Fick's model (Eq. 3), at the optimal values of the identified parameters and more specifically of the moisture diffusivity, is observable in the figures to the left of Fig. 3.

The apparent diffusivities corresponding to the moisture content in the pumpkin cubes at that time point are represented by the moisture diffusivity values computed. The rates of reduced moisture content ratio $MR_{theo}$ are simulated using the identified values of $D_{eff}$ in Eq. (3).

During drying process, as shown in Fig. 3, the instantaneous diffusion coefficients calculated using the Fick model increases with the decrease of the moisture ratio in the first period of the drying process, on the other hand in the second period the instantaneous diffusion coefficients decreases with the increase the moisture ratio. This moisture diffusivity behavior could be the result of a shift in mechanism during the drying process: in the first drying step, the main mass transfer mechanism could be liquid water diffusion. The low results during the initial time could be attributed to the starting superficial interaction, defined as ''starting accessibility,'' reveals the quick removal of humidity from the surface, independently from the diffusion processes.

The following decrease could be attributed to instantaneous diffusion coefficients being dependent on moisture content throughout the drying process. The procedure is also applied to the convective drying data of pumpkin cubes for the experimental design (Table 2). The calculated instantaneous diffusion coefficients values increased and peaked approximately at moisture ratio $MR \approx 0.5$ and then decreased steadily (Fig. 3). The substantial increase in moisture diffusivity at the start of the drying process (i.e. for high water contents) is the most likely due to the preheating of the may be caused by the onset of collapse of the cell structure. Several authors have reported comparable results concerning the moisture diffusion behavior during convective drying of different materials, including apricot pomace (Kayran and Doymaz, 2019), pumpkin fruit leather (Özkan Karabacab *et al*., 2021), pumpkin slices (Benseddik *et al*., 2019) and apple slices (Doymaz and Soydan, 2021). The mean instantaneous diffusion coefficients are estimated between 12.46 x $10^{-9}$ and 63.23 x $10^{-9}$ $m^2/s$ for drying processes optimized by a central composite design to study the effect of three critical variables.

## V. CONCLUSIONS

The drying process of pumpkin cubes was well represented by the proposed approach of identifying the values of instantaneous diffusion coefficient in a bio-logical material using the diffusion equation's serial solution. Moreover, the values of instantaneous diffusion coefficient have been determined, and the drying process of the pumpkin cubes adequately de-scribed by Fick's law of diffusion. It was found that the values obtained varied during the drying process and were de-pendent on the moisture content. The inverse problem of determining the instantaneous diffusion coefficient has been solved using the Levenberg-Marquardt methods. Being statistically validated and applied to eighteen drying experiments according to central composite design, Levenberg-Marquardt optimization methods have shown good performance.

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**Received: February 3, 2022**
**Sent to Subject Editor: March 1, 2022**
**Accepted: August 29, 2022**
**Recommended by Subject Editor Laura Briand**