

# The Effect of External Mass Transfer Resistance during Drying of Fermented Sausage

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The drying mechanism of fermented sausages (sucuks) that were cylindrical rod shaped, 40 cm long and 4 cm diameter, during ripening under natural convection conditions at different temperatures (15 to 30°C) was examined. To simulate the experimental drying curves, three empirical models and a diffusional model assuming negligible external mass transfer resistance were evaluated. The drying rate curves of sucuk samples were also simulated taking into account the influence of the external mass transfer resistance. The equation was solved using the trial-and-error solution algorithm developed in this study and the mass transfer coefficient,  $k_c$ , and effective moisture diffusivity,  $D_{eff}$ , were simultaneously determined ( $1.44 \times 10^{-8}$  to  $1.93 \times 10^{-8}$  m/s and  $4.30 \times 10^{-10}$  to  $6.85 \times 10^{-10}$  m<sup>2</sup>/s, respectively). The proposed model considering the effect of external resistance allowed the accurate simulation of the experimental drying data of sucuks at different temperatures.

**Keywords** Drying models; Effective moisture diffusivity; External resistance; Fermented sausage (sucuk); Ripening process

## INTRODUCTION

Dried-fermented sausage, which is called sucuk, is one of the most popular traditional meat products in Turkey and is produced mainly from beef, beef fat, and tail fat from sheep. To obtain sucuk dough, minced beef and fat are mixed with curing ingredients, garlic paste, and spices (such as black pepper, red pepper, cumin, and allspice), and then sucuk dough is filled into natural or artificial casings. Sucuk is produced in two steps; the first step is the fermentation by added starter culture<sup>[1]</sup> and the second step is the drying of the sucuks under controlled climatic conditions.

Combination of fermentation and drying steps is known as the ripening period. During the ripening step, some physical, microbiological, and biochemical reactions that are responsible for the appearance, flavor, and aroma typical of these products take place<sup>[2]</sup> and the moisture content

of the product decreased as a result of moisture loss at a definite temperature and low RH%.<sup>[3–6]</sup> Ripening periods and temperatures for sucuk change from 6 to 20 days and from 12–14 to 18–20°C, respectively.<sup>[7,8]</sup>

Dehydration during the ripening process contributes to stabilizing the product by decreasing the water activity ( $a_w$ ) value and increases the shelf-life.<sup>[3]</sup> The kinetics of these reactions are affected by the product moisture content, which decreases due to water loss. As a consequence, the dehydration rate during ripening affects the extent of these reactions. Moisture transfer from the meat product to the ambient air during the drying process depends on its moisture content and on its composition. Nevertheless, process conditions such as temperature, velocity, and relative humidity (RH) of ambient air and characteristics of the boundary layer are also important.<sup>[2,3]</sup>

Studies dealing with the modeling of the drying process of meats and meat products have received increasing attention in recent years.<sup>[2,3,9–15]</sup> Some are related with the drying mechanism and the ripening conditions of fermented meat products, especially sucuk. Simal et al.<sup>[2]</sup> examined the drying mechanism of traditional meat-based product (sobrassoda) during ripening and determined effective diffusion coefficient for water loss as  $2.86 \times 10^{-11}$  m<sup>2</sup>/s at 14°C and 85% RH taking into account the external mass transfer resistance. Gou et al.<sup>[13]</sup> also determined the effective water diffusivity in dry cured ham as  $10^{-11}$  m<sup>2</sup>/s at 13°C. Trujillo et al.<sup>[14]</sup> examined drying of beef meat at different temperatures (6.8–40.4°C) and relative humidities (78–92%, RH) and calculated the effective water diffusivity in beef meat as  $10^{-10}$  m<sup>2</sup>/s, using different drying models.

The main objective of this study was to examine the drying mechanism of sucuk during ripening at different temperatures (15, 20, 25, and 30°C), evaluate alternate empirical or simple phenomenological models reported in the literature to simulate the drying curves of sucuk, and propose a model to accurately simulate the drying kinetics of sucuk, taking into account the external mass transfer resistance during ripening at different temperatures.

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## MATHEMATICAL MODELING

The mechanism of mass transfer in foods is complex. Frequently, the modeling of the drying process during the falling rate period is carried out by assuming that the main mechanism is of diffusional nature.<sup>[16]</sup> That is, it is assumed that the moisture driving force during drying is a liquid concentration gradient. The effect of heat transfer is neglected since the heat transfer proceeds in a rapid manner during drying. Furthermore, it is assumed that the diffusion coefficient of moisture is the same in all directions (isotropic material) and shrinkage of sample is negligible. Under these conditions, moisture transfer from the solid in the falling rate period can be described by unsteady-state Fick's law of diffusion:

$$\frac{\partial X(r,t)}{\partial t} = D_{eff} \left( \frac{\partial^2 X(r,t)}{\partial r^2} + \frac{1}{r} \frac{\partial X(r,t)}{\partial r} \right) \quad (1)$$

The solution for Eq. (1) in series, for initial condition of uniformly distributed moisture throughout the solid (Eq. (2)), boundary conditions of central symmetry of the solid material (Eq. (3)), and convective boundary at the surface (Eq. (4)) is given for an infinite cylinder geometry in Eq. (5).

$$X(r,t)|_{t=0} = X_0 \quad (2)$$

$$\left. \frac{\partial X(r,t)}{\partial r} \right|_{r=0} = 0 \quad (3)$$

$$-D_{eff} \cdot \rho_{dm} \left. \frac{\partial X(r,t)}{\partial r} \right|_{r=R} = k_c (\varphi_s - \varphi_a) \quad (4)$$

$$\begin{aligned} \psi(r,t) &= \frac{X(r,t) - X_e}{X_0 - X_e} \\ &= \sum_{n=1}^{\infty} \left[ \frac{2 J_1(\lambda_n)}{\lambda_n J_0^2(\lambda_n) + J_1^2(\lambda_n)} J_0\left(\lambda_n \frac{r}{R}\right) \exp\left(-\lambda_n^2 \frac{D_{eff} t}{R^2}\right) \right] \end{aligned} \quad (5)$$

where  $\lambda_n$  are the roots of Eq. (6):

$$Bi = \frac{k_c R}{D_{eff}} = \lambda_n \frac{J_1(\lambda_n)}{J_0(\lambda_n)} \quad (6)$$

Equation (5) gives the moisture concentration at a given time and a given location in an infinite cylinder. Since the experimental data were obtained for total moisture loss through the whole product, Eq. (5) integrated throughout the whole volume to result in Eq. (7) to enable the use of experimental data. Equation (7) gives the average moisture concentration in the whole sample as a function of time.

$$\bar{\psi} = \frac{\bar{X}_t - X_e}{X_0 - X_e} = \sum_{n=1}^{\infty} \left[ \frac{4 J_1^2(\lambda_n)}{\lambda_n^2 J_0^2(\lambda_n) + J_1^2(\lambda_n)} \exp\left(-\lambda_n^2 \frac{D_{eff} t}{R^2}\right) \right] \quad (7)$$

To determine the effective diffusion coefficient ( $D_{eff}$ ), infinite and finite mass transfer coefficient approaches may be applied. In analysis of the drying mechanism, an infinite external convective mass transfer coefficient, therefore an infinite Biot number approach has been widely used in the literature.<sup>[17,18]</sup> However, especially under the natural convection conditions (due to low air velocity), the negligible external mass transfer resistance approach may not be true and may result in inaccurate results.<sup>[2,19-22]</sup> Considering the effect of external resistance is important, Eq. (7) can be used to determine  $D_{eff}$ .

For the case of negligible external convective mass transfer resistance, therefore an infinite Biot number, the roots of Eq. (6),  $\lambda_n$  values are determined from  $J_0(\lambda_n) = 0$ . By applying the infinite external convective mass transfer approach, Eq. (7) can be rewritten as Eq. (8):

$$\bar{\psi} = \frac{\bar{X}_t - X_e}{X_0 - X_e} = \sum_{n=1}^{\infty} \left[ \frac{4}{\lambda_n^2} \exp\left(-\lambda_n^2 \frac{D_{eff} t}{R^2}\right) \right] \quad (8)$$

For sufficiently long drying times, the change in dimensionless moisture content becomes linear, which allows the calculation of  $D_{eff}$  from the slope of the dimensionless moisture content vs. drying time curve by using the only first terms ( $n=1$ ) of Eqs. (7) and (8).<sup>[23]</sup> This approach is based on a constant diffusion coefficient. If the numerical value for the  $k_c$  was known, Eq. (6) may be used to determine  $\lambda_1$  value, then leads to the value of Biot number and therefore the  $D_{eff}$  through Eq. (7). The mathematical models explained above to determine  $D_{eff}$  based on the unsteady-state Fick's law of diffusion developed by Crank.<sup>[24]</sup>

Besides the theoretical models, several researchers have proposed quite simple models to simulate the drying curves of food that can provide adequate representation of experimental data although the parameters of these models lack physical sense.<sup>[25]</sup> Among semi-empirical drying models, namely the Henderson and Pabis model (Eq. (9)), the Lewis or exponential model (Eq. (10)) and the Page model (Eq. (11)) are widely used.<sup>[18,25-28]</sup>

$$\frac{\bar{X}_t - X_e}{X_0 - X_e} = a \exp(-kt) \quad (9)$$

$$\frac{\bar{X}_t - X_e}{X_0 - X_e} = \exp(-kt) \quad (10)$$

$$\frac{\bar{X}_t - X_e}{X_0 - X_e} = \exp(-kt^n) \quad (11)$$

where  $k$  is the drying rate constant (1/s) or (1/s<sup>n</sup>) and  $a$  and  $n$  are parameters in the models.

**MATERIALS AND METHODS**

**Materials**

Fresh boneless beef cuts (80%) and beef fat (20%) obtained from a local manufacturer in Izmir, Turkey, were used as raw materials. The following ingredients were added per kilogram of meat mixture, 27.5 g spices, 10 g garlic, 28 g curing ingredients, and 0.5 g lyophilized starter culture mixture (*Staphylococcus xylosus* + *Pediococcus acidilactici*). The meat was ground in a 3-mm-diameter meat grinder, then the curing ingredients and spice mix were added and mixed to obtain homogenous sucuk dough. Sucuk dough was filled using a hydraulic sausage filling machine (Alpina-SG, Schwei, Germany) into artificial collagen casings, and cylindrical rod-shaped sucuks with a length of 40 cm and a diameter of 4 cm were obtained. Sucuks were then hung on stainless steel hangers and placed in a drying chamber for the ripening process.

**Experimental Procedure**

The ripening process (drying) was carried out in a laboratory-scale controlled chamber at a constant air velocity (0.5 m/s) for 7 days. The dimensions of the drying chamber were 0.83 m × 0.64 m × 62 m. The chamber was equipped with small ventilators to ensure the correct distribution of the air. The drying air was supplied from the atmosphere without regulating humidity. Average relative humidity (RH) of the air inside the drying chamber varied between 65 and 85% RH during the experiments. Four different air temperatures (15, 20, 25, and 30°C) were used for the ripening process.

The ripening process was continued within 7 days. During this period, two sucuks from each batch were removed on 0 (initial), 1st, 2nd, 3rd, 4th, 5th, 6th, and 7th days of ripening and moisture content and water loss of samples were analyzed. All determinations were carried out in duplicate. The average moisture content of sucuk samples was determined using the oven method at 105 ± 2°C.<sup>[29]</sup> Water loss during drying was measured by weighing the product using an electronic balance (Gec-Avery Berkel model no: CB062, West Midlands, England) with an accuracy of 0.01 g. The process was replicated under the same conditions to ensure uniformity of the results.

**Data Analysis**

Analysis of the experimental data and statistical modeling were performed using linear and nonlinear regression analysis (MS Excel, 2003). The Henderson-Pabis model, the Lewis model (exponential model), and the Page model were fitted to the experimental drying data. In order to evaluate the goodness of the fit of the tested models to the experimental data, the coefficient of determination ( $R^2$ ), reduced chi-square ( $\chi^2$ ), and root mean square error (RMSE) were used as criteria. The best model describing

the drying behavior was chosen as the one with the highest  $R^2$  and the least  $\chi^2$  and RMSE.<sup>[26,30,31]</sup>

These parameters can be described by Eqs. (12) and (13) as

$$\chi^2 = \frac{\sum_{i=1}^N (\bar{\Psi}_{exp_i} - \bar{\Psi}_{pre_i})^2}{N - Z} \tag{12}$$

$$RMSE = \left[ \frac{1}{N} \sum_{i=1}^N (\bar{\Psi}_{exp_i} - \bar{\Psi}_{pre_i})^2 \right]^{1/2} \tag{13}$$

where  $\bar{\Psi}_{exp_i}$  is the experimental dimensionless moisture value,  $\bar{\Psi}_{pre_i}$  is the predicted dimensionless moisture value from the model,  $N$  is the number of observations, and  $Z$  is the number of constants.

**RESULTS AND DISCUSSION**

Drying curves were studied during 7 days of ripening for the average moisture contents varying from 1.8 to 0.5 kg water/kg DM for the drying set experiments. The variation of the average moisture content of sucuk samples with drying time is shown in at different drying temperatures. The standard deviation of the each experimental point for moisture content of the samples ranges between 0.020 and 0.094 kg water/kg DM.

Although the initial moisture contents of the samples were quite high, a constant drying rate period was not observed under the experimental conditions employed and the overall drying process took place in the falling rate period. As seen in Fig. 1, an increase in the temperature promoted a significantly increase in water loss. As expected, air temperature affected the drying curves, decreasing the drying time of samples.

The equilibrium moisture content,  $X_e$ , of the samples at the temperatures studied was determined experimentally by keeping the samples in the drying chamber for a one-week

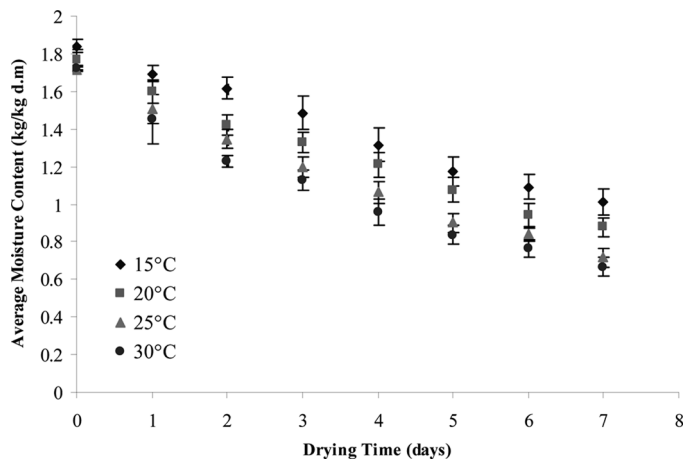


FIG. 1. Changes in moisture content of the sucuk samples during drying at different temperatures.

TABLE 1  
Parameters of the three semi-empirical drying models for drying kinetics of sucuk samples

Model	Model parameters/ goodness of fit	Drying temperature (°C)			
		15	20	25	30
Henderson and Pabis	$k(s^{-1})$	$1.385 \times 10^{-6}$	$1.515 \times 10^{-6}$	$1.787 \times 10^{-6}$	$1.864 \times 10^{-6}$
	$a$	1.0527	1.0153	1.0162	0.9609
	$R^2$	0.9882	0.9916	0.9946	0.9954
	$\chi^2$	0.0005	0.0003	0.0002	0.0002
	RMSE	0.0203	0.0149	0.0130	0.0115
Exponential	$k(s^{-1})$	$1.247 \times 10^{-6}$	$1.473 \times 10^{-6}$	$1.741 \times 10^{-6}$	$1.983 \times 10^{-6}$
	$R^2$	0.9648	0.9900	0.9934	0.9916
	$\chi^2$	0.0009	0.0003	0.0002	0.0003
	RMSE	0.0272	0.0157	0.0139	0.0166
Page	$k(s^{-n})$	$7.512 \times 10^{-8}$	$6.400 \times 10^{-7}$	$8.078 \times 10^{-7}$	$5.707 \times 10^{-6}$
	$n$	1.2171	1.0640	1.0596	0.9177
	$R^2$	0.9867	0.9930	0.9912	0.9953
	$\chi^2$	0.0003	0.0002	0.0002	0.0002
	RMSE	0.0158	0.0138	0.0116	0.0116

period to reach constant weight. The equilibrium moisture contents of the samples were found to be in the range of 0.1919 and 0.3231 kg/kg DM, which is in accordance with the findings of Simal et al.<sup>[2]</sup> and Lomauro et al.<sup>[32]</sup> for meat-based products.

### Evaluation of the Empirical Models

Experimental results of dimensionless moisture content ( $\bar{X}_t - X_e / X_0 - X_e$ ) with drying time were fitted to the proposed semiempirical models; namely, the Henderson and Pabis model (Eq. (9)), the exponential model (Eq. (10)), and the Page model (Eq. (11)). By using the solver, an optimization tool (GRG2 method) included in the Microsoft Excel (2003) spreadsheet, the parameters of the different models that provided the lowest sum of square differences between the experimental and the predicted moisture values were identified. Table 1 shows the parameters of the models and also the criteria ( $R^2$ ,  $\chi^2$ , RMSE) for the models.

From the values of coefficient of determination ( $R^2$ ), the reduced chi-square ( $\chi^2$ ), and the root mean square error (RMSE), it is clear that all models gave satisfactorily good fit in predicting the moisture content of sucuk samples during drying. The  $R^2$  values for the three models were always greater than 0.95, and in all cases, the  $\chi^2$  values and the RMSE were less than 0.0115 and 0.0272, respectively.

For all empirical models, the drying rate constant,  $k$ , increased with drying temperature. The identified parameters  $k$  and  $n$  of the Page model are also shown in Table 1. Although the results for constant  $n$  value with the drying temperature were obtained by Senadeera et al.<sup>[33]</sup> and Simal et al.<sup>[25]</sup> in this study, the estimated figure for the  $n$  parameter of the Page model did not stay

constant with the drying temperature. Comparisons of the experimental and predicted dimensionless moisture contents obtained using the three empirical models for 15 and 30°C are shown in Fig. 2. As can be observed, a more accurate simulation of the drying curves at higher drying temperatures was obtained using the Henderson and Pabis model.

### Determination of Moisture Diffusivity in Case of Negligible External Resistance

During the studied ripening (drying) period, the shrinkage effects were ignored. Therefore, the diffusional model

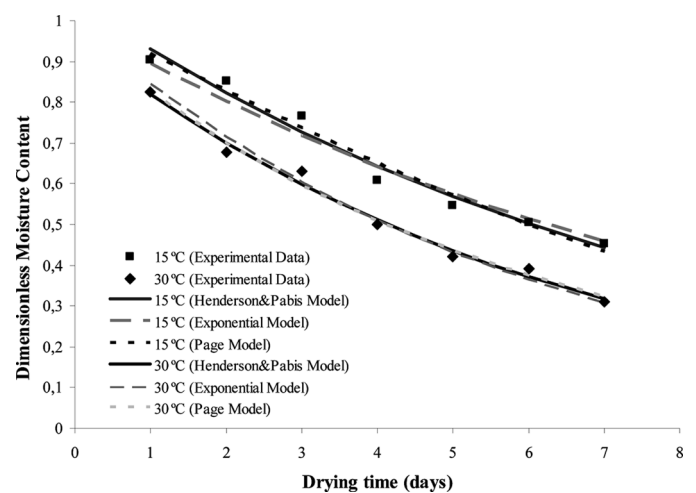


FIG. 2. Experimental and predicted drying curves of sucuk samples at 15 and 30°C. Simulation obtained by using the three proposed empirical models.

TABLE 2

Effective moisture diffusivity and mass transfer coefficient proposed for simulating the drying curves of sucuk samples and the criteria for the models

Hypothesed diffusional model	Model parameters/ goodness of fit	Drying temperature (°C)			
		15	20	25	30
Negligible External Resistance (NER)	$D_{eff}(m^2/s)$	$3.48 \times 10^{-11}$	$4.61 \times 10^{-11}$	$5.99 \times 10^{-11}$	$7.31 \times 10^{-11}$
	$R^2$	0.5876	0.7041	0.7269	0.8420
	$\chi^2$	0.0069	0.0051	0.0055	0.0031
	RMSE	0.0519	0.0687	0.0659	0.0766
Considering External Resistance (CER)	$D_{eff}(m^2/s)$	$4.30 \times 10^{-10}$	$4.72 \times 10^{-10}$	$5.59 \times 10^{-10}$	$6.85 \times 10^{-10}$
	$k_c(m/s)$	$1.44 \times 10^{-8}$	$1.56 \times 10^{-8}$	$1.85 \times 10^{-8}$	$1.93 \times 10^{-8}$
	$R^2$	0.9728	0.9803	0.9916	0.9901
	$\chi^2$	0.0006	0.0003	0.0002	0.0006
	RMSE	0.0223	0.0155	0.0138	0.0113

was solved assuming negligible shrinkage and adapting the initial dimensions. When external resistance to mass transfer is considered negligible, the average moisture content can be calculated, according to the proposed model, from Eq. (8). If the average moisture contents at different time intervals, equilibrium moisture content and roots of the transcendental equation (Eq. (6)) when Biot number goes to infinity are known, the effective diffusion coefficient can be identified using Eq. (8) by minimize the differences between experimental and estimated dimensionless moisture contents. For the parametric identification, SOLVER, the above-mentioned tool of the Excel spreadsheet, was used. Sufficient terms of the infinite series solution in Eq. (8) were used in order to achieve an error lower than 1%.

The predicted values for the effective moisture diffusivity ( $D_{eff}$ , Table 2) during ripening of sucuk samples increased with temperature and were found to be in the range of  $3.48 \times 10^{-11}$  to  $7.31 \times 10^{-11} m^2/s$ . These are similar to the effective diffusivities, determined using a negligible external resistance approach, proposed by different authors for other meat products:  $1.1 \times 10^{-11} m^2/s$  in pork loin at 10°C,<sup>[34]</sup> from  $6.45$  to  $9.28 \times 10^{-11} m^2/s$  at 13°C,<sup>[13]</sup> and from  $2.65$  to  $3.71 \times 10^{-11} m^2/s$  at 5°C<sup>[35]</sup> in muscles of pork ham and  $1.07 \times 10^{-11} m^2/s$  at 14°C in meat-based product.<sup>[2]</sup> Besides, Trujillo et al.<sup>[14]</sup> reported a higher diffusivity value of  $1.29 \times 10^{-10} m^2/s$  at 6.8°C for beef at zero surface resistance.

The temperature dependence of the effective diffusion coefficient was described by the Arrhenius-type relation (Eq. (14), Fig. 3). The activation energy,  $E_a$ , was found to be 29.7 kJ/mol for the case of negligible external resistance. A similar  $E_a$  value (28.1 kJ/mol) was found by Trujillo et al.<sup>[14]</sup>

$$D_{eff} = 9.642 \times 10^{-6} \times e^{[-3579/T(k)]} (R^2 = 0.992) \quad (14)$$

The percentage of explained variance obtained by comparing the experimental dimensionless moisture contents and those predicted by the proposed model for all drying conditions ranged between 58.76 and 84.20%. Also, the differences between experimental and estimated dimensionless moisture contents for sucuk samples can be seen in Fig. 4. From these results, it was concluded that hypotheses of negligible external resistance to mass transfer for solving the differential equation representative of the mass transport (Eq. (8)) might not be adequate for the studied system under natural convection conditions.

**Simultaneous Identification of Mass Transfer Coefficient and Effective Moisture Diffusivity Considering the Effect of External Resistance**

When drying air velocity and drying rate are low, as in sucuk drying, the external resistance to mass transport

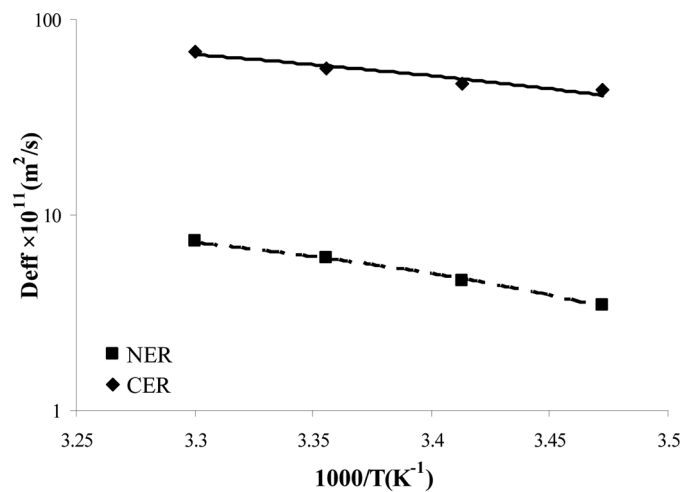


FIG. 3. Arrhenius plot of effective moisture diffusivities. NER: negligible external resistance; CER: considering the effect of the external resistance.

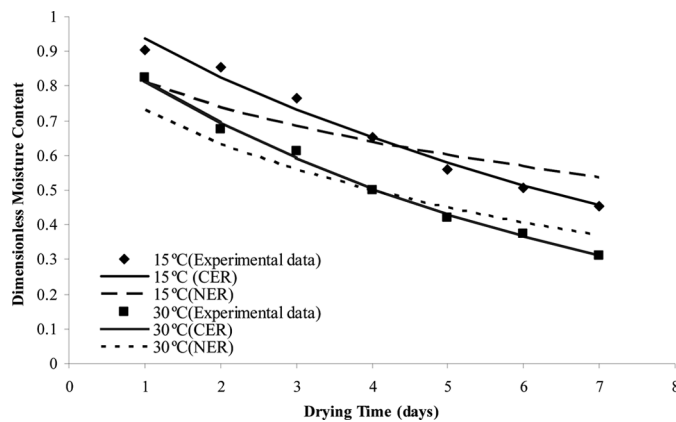


FIG. 4. Estimated and experimental dimensionless moisture content vs. drying time at temperatures of 15 and 30°C. Simulation considering negligible external resistance (NER) and the effect of the external resistance (CER).

frequently becomes important and cannot be neglected. In order to take into account both external and internal resistances, Eq. (1) can be solved by assuming the boundary conditions given in Eqs. (2), (3), and (4). Thus, the effective moisture diffusivity could be estimated during drying by using Eqs. (6) and (7) for average moisture content as long as the mass transfer coefficient is known. The problem in using these equations is the adequate estimation of the mass transfer coefficient. Several attempts have been made to determine the mass transfer coefficient during drying.<sup>[2,22,23,36–39]</sup>

In drying processes, the general approach is to assume an infinite mass transfer coefficient and infinite Biot number and then to determine the effective diffusion coefficient easily from the series solution of Fick's second law of diffusion, neglecting external resistance. It is a general simplifying assumption that has been used numerous times in the literature.<sup>[16,17,40–43]</sup> However, especially under natural convection conditions and at low temperatures, this approach might lead to incorrect results.

Although numerous studies have been conducted to determine effective diffusivities of food products subjected to drying, the literature provides scant information on the topic of convective mass transfer coefficients. For example, Fahloul et al.<sup>[44]</sup> determined a finite value of mass transfer coefficient using the vapor pressure difference as the driving force. Use of the heat-mass convection analogies, i.e., the Chilton-Colburn analogy, is another approach to determine the mass transfer coefficient when the heat transfer coefficient is known.<sup>[20,22]</sup> Markowski<sup>[36]</sup> developed a methodology to determine the mean value of mass transfer coefficient based on the mass average change of moisture undergoing a simultaneous heat and mass transfer process. According to this method, the mass transfer coefficient was determined using the weight loss data obtained to represent

the average mass change instead of the changes that have been occurring on the surface. Another way to estimate the value of mass transfer coefficient may be the use of the humidity ratio difference between the surface and the medium as the driving force for the mass transfer.<sup>[45]</sup> Bialobrzeski and Markowski<sup>[21]</sup> and Bialobrzeski<sup>[37]</sup> reported that the experimental data on time-related changes in moisture content during drying can be used to simultaneously determine the mass transfer coefficient and effective moisture diffusivity, based on an inverse problem formulation approach.

Simultaneous determination of both effective diffusion coefficient and external mass transfer coefficient parameters can be achieved with less effort as compared to the methods discussed above by developing a trial-and-error solution algorithm as given in Fig. 5. This algorithm is based on the minimization of the residual sum of squares between experimental and estimated average dimensionless moisture contents by using the equation that is a

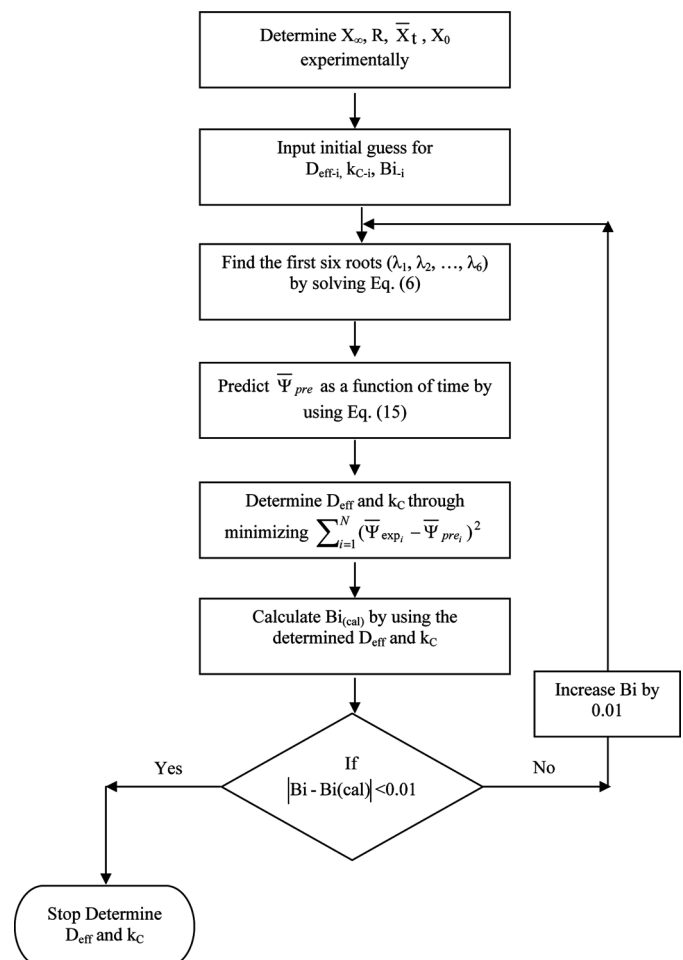


FIG. 5. Algorithm for trial-and-error solution to determine mass transfer coefficient and effective moisture diffusivity simultaneously.

combination of Eqs. (6) and (7), given below:

$$\bar{\psi} = \frac{X_t - X_e}{X_0 - X_e} = \sum_{n=1}^{\infty} \left[ \frac{4 \left( \frac{k_c J_0(\lambda_n R)}{D_{eff} \lambda_n} \right)^2}{\lambda_n^2 \left( \frac{D_{eff} J_1(\lambda_n R)}{k_c} \right)^2 + \left( \frac{k_c J_0(\lambda_n R)}{D_{eff} \lambda_n} \right)^2} \exp\left(-\lambda_n^2 \frac{D_{eff} t}{R^2}\right) \right] \quad (15)$$

Equation (15) allows the simultaneous estimation of mass transfer coefficient and effective diffusion coefficient if the characteristic roots ( $\lambda_n$ ) of the transcendental equation are known. Therefore, a spreadsheet programme in Visual Basic for Excel was written to solve the equations and to identify both the  $D_{eff}$  and  $k_c$  parameters. At the first step, an initial guess has been made for  $D_{eff}$ ,  $k_c$ , and Biot number, and the right-hand side of Eq. (6) was solved for the first six roots by using the Newton-Raphson method. Estimated average dimensionless moisture contents ( $\bar{\psi}_{pre}$ ) are calculated with these roots and the initial guess for  $D_{eff}$  and  $k_c$  utilizing Eq. (15). Then, the Nelder-Mead downhill simplex algorithm, which is a popular derivative-free optimization method, was used to minimize the residual sum of squares between experimental and estimated average dimensionless moisture contents by changing both  $D_{eff}$  and  $k_c$ . Therefore, applying the algorithm in Fig. 5, the effective diffusivity and mass transfer coefficient values were simultaneously determined. The identified effective diffusivity and mass transfer coefficient values with  $R^2$ ,  $\chi^2$ , and RMSE values at different temperatures are also given in Table 2.

Recent available literature provide many values of the mass transfer coefficient for food products. Mikenatic et al.<sup>[46]</sup> reported that during drying a layer of barley at 75°C and at an air velocity of 0.56 m/s,  $k_c$ , evaluated from analogy between heat and mass transfer, was  $1.08 \times 10^{-6}$  m/s. Markowski<sup>[36]</sup> determined  $k_c$  as  $1.37 \times 10^{-7}$  m/s at 60°C under natural convection drying of carrots. Martynenko<sup>[39]</sup> determined that the convective mass transfer coefficients were  $0.2 \times 10^{-7}$  m/s at 38°C and  $0.45 \times 10^{-7}$  m/s at 50°C at low air velocity for ginseng root drying. Carcel et al.<sup>[38]</sup> estimated mass transfer coefficient and effective diffusion coefficient as  $0.54 \times 10^{-3}$  kg water/m<sup>2</sup>s and  $5.25 \times 10^{-10}$  m<sup>2</sup>/s, respectively, for persimmon drying at low air velocity, using the model taking external resistance into account. Bon et al.<sup>[22]</sup> also showed that the effect of the external resistance on mass transfer curves was important and they determined mass transfer coefficient as  $7.7 \times 10^{-4}$  kg water/m<sup>2</sup>s at 50°C and  $1.6 \times 10^{-3}$  kg water/m<sup>2</sup>s at 90°C (air velocity 1.1 m/s) for apricot drying. For meat products, Simal et al.<sup>[2]</sup> found that  $k_c$  was  $3.99 \times 10^{-3}$  m/s during ripening of meat-based product at 14°C and 85% RH under natural convection conditions, whereas under similar conditions, we found  $k_c$

values in the range of  $10^{-8}$  m/s and smaller than those of Simal et al.<sup>[2]</sup> When the Biot number was calculated using the values of Simal et al.,<sup>[2]</sup> it took a great valued and approached infinity. Hence, the authors' decision about the influence of the external resistance on moisture transfer should not be valid. The other  $k_c$  values given in the literature were in an acceptable range, although they were obtained for different conditions.

As can be observed in Table 2, the identified  $D_{eff}$  and  $k_c$  showed an increasing trend as the drying temperature increased. In the case of significant external resistance,  $D_{eff}$  was found to be considerably higher than  $D_{eff}$  when the external resistance was neglected. The temperature dependence of  $D_{eff}$  was described according to the Arrhenius-type relation (Fig. 3). The activation energy,  $E_a$ , was found to be 23.8 kJ/mol. A similar relationship among  $D_{eff}$ ,  $k_c$ , and temperature was found by Bon et al.<sup>[22]</sup> and Trujillo et al.<sup>[14]</sup>

$$D_{eff} = 8,456 \times 10^{-6} \times e^{[-2860/T(k)]} (R^2 = 0.995) \quad (16)$$

In order to evaluate the accuracy of the proposed model solved by considering the influence of the external resistance to mass transfer, the drying curves at 15 and 30°C were simulated by using simultaneously identified  $D_{eff}$  and  $k_c$ , as seen in Fig. 4. It can be concluded that the proposed model solved by taking into account the external resistance provided a considerably better correlation with the experimental data, so the simulation was more accurate when both  $k_c$  and  $D_{eff}$  were identified in case of significant external resistance. The similar approach taking into account of the convective mass transfer resistance at low temperature and low air velocity drying was also proposed

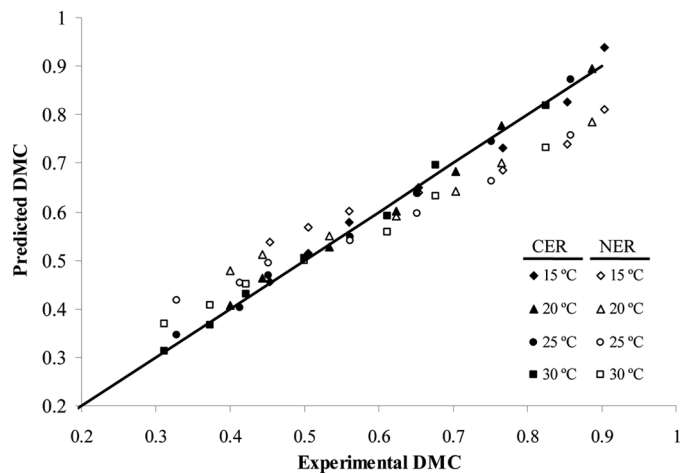


FIG. 6. Comparison of the experimental and predicted dimensionless moisture content (DMC). Simulation obtained by using the proposed model considering external resistance (CER) and negligible external resistance (NER).

earlier by Simal et al.<sup>[2]</sup> and Bon et al.<sup>[22]</sup> The model takes into account the shrinkage of the meat samples was proposed by Trujillo et al.<sup>[14]</sup>

The experimental versus predicted dimensionless moisture content (DMC), which is obtained by using the models considering external resistance (CER) and negligible external resistance (NER), for drying at four different temperatures are plotted in Fig. 6. As can be observed in Fig. 6, the best fit was obtained when the model was solved by taking into account the effect of external resistance.

## CONCLUSION

The effect of the temperature on the drying (ripening) process of fermented-type sausages (sucuks) has been observed. The proposed diffusional model solved by considering the influence of the external resistance to mass transfer gave the accurate simulation of the drying curves of sucuk samples at low air velocities, whereas the diffusional model solved by neglecting the external resistance was found to give inaccurate results. The latter was not adequate to determine  $D_{eff}$  under natural convection conditions. For simultaneous determination of mass transfer coefficient and effective diffusivity, the trial-and-error solution algorithm was developed and this solution gave considerably accurate results. The applicability of the proposed model to simulate the drying curves during ripening of meat products or drying of food products under natural convection conditions could be expected. Although the empirical models do not provide mechanistic information and the estimated parameters have no physical meaning, it is possible to accurately simulate the drying curves, which, from a practical point of view could be quite interesting.

## NOMENCLATURE

$a, n$	Parameters in models in Eqs. (9), (11)
$Bi$	Biot number, $Bi = \frac{k_c R}{D_{eff}}$
$D_{eff}$	Effective diffusion coefficient ( $m^2/s$ )
$E_a$	Activation energy (kJ/mol)
$J_0, J_1$	The first kind, 0th-and 1st-order Bessel functions
$k$	Parameter in models in Eqs. (9), (10) ( $s^{-1}$ )
$k$	Parameter in model in Eq. (11) ( $s^{-n}$ )
$k_c$	Convective mass transfer coefficient (m/s)
$N$	Number of observations (Eqs. (12) and (13))
$R$	Radius of the cylinder (m)
$R^2$	Coefficient of determination
RMSE	Root mean square error (Eq. (13))
$r$	Distance from center (m)
$T$	Temperature (K or °C)
$t$	Time (s)
$X_0$	Initial moisture content (kg water/kg dry matter)
$X_e$	Equilibrium moisture content (kg water/kg dry matter)
$X_{(r,t)}$	Local moisture content (kg water/kg dry matter)

$\bar{X}_t$	Average moisture content (kg water/kg dry matter)
$Z$	Number of constants in the models (Eq. (12))

## Greek Letters

$\lambda_n$	Root of Eq. (6)
$\rho_{dm}$	Dry matter density (kg dry matter/ $m^3$ )
$\varphi_a$	Medium moisture concentration (kg water/ $m^3$ )
$\varphi_s$	Moisture concentration in the solid surface (kg water/ $m^3$ )
$\chi^2$	Chi-square (Eq. (12))
$\bar{\psi}$	Average dimensionless moisture content
$\bar{\psi}_{exp}$	Experimental average dimensionless moisture content
$\bar{\psi}_{pre}$	Predicted average dimensionless moisture content
$\psi_{(r,t)}$	Local dimensionless moisture content

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