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Research article

DEVELOPMENT OF A MATHEMATICAL MODEL TO DETERMINE THE MOISTURE DIFFUSIVITY OF AN INFINITE FLAT SLAB DRYING MATERIAL: APPLICATION TO FREEZE-DRYING OF YOGURT

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ABSTRACT
In the study of freeze-drying engineering and technology, one of the key factors is the development and solution of a mathematical modeling problem to describe the drying kinetics, thereby determining the optimal technological parameters. The ultimate goal is to ensure that the dried product meets high quality standards, achieves the desired moisture content, and minimizes energy consumption. However, this modeling and optimization process cannot be effectively carried out without first determining the characteristic moisture diffusion coefficient of the material. In particular, the characteristic moisture diffusivity coefficient of the material depends on the temperature and pressure of the freeze-drying environment, which, until now, has not been addressed in any research work. Therefore, this study presents a mathematical model for determining the effective moisture diffusivity of yogurt during the freeze-drying process: $D = (0.284 + 0.05\times X_1 + 0.123\times X_2 + 0.144\times X_1\times X_2)\times 10^{-11}$, m²/s, with the maximum error of 1.11% for the mathematical model and the correlation coefficient $R^2 = 0.9979$. Based on this model, when the freeze-drying temperature $X_1 = T = (30\div40)^0C$ and the pressure $X_2 = P = (0.01\div0.1)$mmHg the effective moisture diffusivity of yogurt during freeze-drying was determined to range from 1.822×10^{-11} to 2.864×10^{-11} m²/s.

1. Introduction

Over the past two decades, freeze-drying technology and engineering have developed rapidly worldwide and are now considered to have reached a high level of maturity (Fellows, 2000; Dzung et al., 2022). This advancement has made significant contributions to the development of various fields such as pharmaceuticals, food processing, biotechnology, and related industries.

Freeze-drying is conducted under low-pressure and low-temperature conditions, specifically below the triple point of water O (0.0098∘C; 4.58 mmHg). This means that the temperature of the drying material remains below 0.0098∘C and the ambient drying pressure stays below 4.58 mmHg. As a result, the dried product retains almost all of the original natural characteristics of the raw material: proteins are neither hydrolyzed nor denatured, carbohydrates are not hydrolyzed or gelatinized, lipids are not oxidized; color and flavor are preserved; bioactive compounds and vitamins remain intact; and trace minerals are conserved (Anandharamakrishnan, C., 2017; Dzung, N.T and et al., 2024).

In addition, the final product structure is porous, with no shrinkage or surface cracking, and exhibits excellent rehydration capability—superior to that of products dried using conventional methods (Bhushani & Anandharamakrishnan, 2017; Athanasios, I. Liapis and Roberto Bruttini, 2020; Dzung et al., 2022). Therefore, freeze-drying is considered one of the most advanced drying technologies available today. Many years of experimentation have shown that freeze-drying is highly suitable for drying yogurt.

Yogurt is a dairy product fermented mainly by lactic acid bacteria such as Lactobacillus delbrueckii subsp. bulgaricus and Streptococcus thermophilus. Nutritionally, yogurt contains a high level of easily digestible proteins, calcium, B vitamins (particularly B2 and B12), as well as various essential minerals. Importantly, during the fermentation process, lactose in milk is converted into lactic acid, making yogurt more tolerable for individuals with lactose intolerance (Dzung et al., 2024).

In addition to its nutritional value, yogurt is also considered a probiotic food. Beneficial bacteria in yogurt, such as Lactobacillus and Bifidobacterium, can help balance the gut microbiota, inhibit the growth of harmful microorganisms, and stimulate the immune system. Numerous clinical studies have demonstrated that the consumption of probiotic yogurt improves digestion, enhances nutrient absorption, reduces the risk of gastrointestinal disorders, and supports overall health, (Dzung et al., 2024).

The studies by Dzung et al. (2024) have demonstrated that no drying method other than freeze-drying can preserve the structure, nutritional composition, and probiotic content of yogurt.

Due to the high energy costs associated with the freeze-drying process, this method is typically suitable only for raw materials with high economic value. Therefore, it is essential to optimize the drying process to ensure that the final product achieves the highest possible quality, meets the required moisture content for extended shelf life and marketability, and minimizes energy consumption. Only under such conditions can freeze-drying provide a competitive advantage (Athanasios I. Liapis and Roberto Bruttini, 2020; Mawilai, P., Chaloeichitratham, N., and Pornchaloempong, P., 2019).

However, the modeling and optimization of the freeze-drying process are far from straightforward. This is not only due to the inherently complex nature of simultaneous heat and mass transfer phenomena, but also because the effective moisture diffusivity of the drying material—a critical parameter required to solve the mathematical model—has not yet been determined (G. Wilhelm-Oetjen and Peter Haseley, 2004; Bhushani and Anandharamakrishnan, 2017; Dzung et al., 2022).

To date, numerous studies have reported the determination of effective moisture diffusivity for various types of drying materials, such as: Effective moisture diffusivity of plain yogurt undergoing microwave vacuum drying (Suk Shin Kim, Santi R. Bhowmik, 1995); Dehydration characteristics of papaya (Carica pubenscens): determination of equilibrium moisture content and diffusion coefficient (R. Lemus-Mondaca et al., 2007); Dependence of the effective diffusion coefficient of moisture on thickness and temperature in convective drying of sliced materials. A study on slices of banana, cassava and pumpkin (W.J.N. Fernando, H.C. Low, A.L. Ahmad, 2011). However, the specific determination of moisture diffusivity in frozen yogurt in the form of flat slabs—intended for application in freeze-drying—has not yet received sufficient attention (Lim, Y., Hong, S., Shin, Y. K. Kang, S. H., and Center D., 2015). Therefore, the objective of this study is to develop a mathematical model for determining the effective moisture diffusivity of drying materials in general, and frozen yogurt slabs in particular. The findings of this research will serve as a basis for the modeling and optimization of the freeze-drying process (Bhushani and Anandharamakrishnan, 2017; Dzung N.T., Chuyen H.V., Linh V.T.K., et al., 2022; Liviu Giurgiulescu, Phong Le Thanh, Linh T.K. Do, and Tan Dzung Nguyen., 2024).

2. Development of a Mathematical Model for Freeze-Drying of Yogurt

2.1. Initial Assumptions for Developing the Mathematical Model

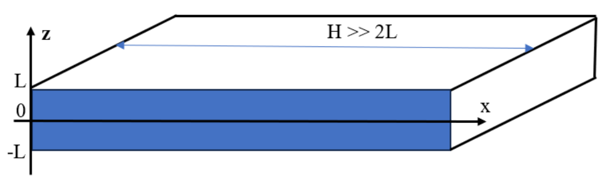

Figure 1a. Physical model of frozen yogurt in flat slab form

Frozen yogurt is cast into flat slab molds with length H (m), width h (m), and thickness a = 2L (m). Since H and h are much larger than the thickness a = 2L (i.e., H, h ≫ a = 2L), the flat slab can be considered as infinite in the length and width directions, (Dzung et al., 2022).

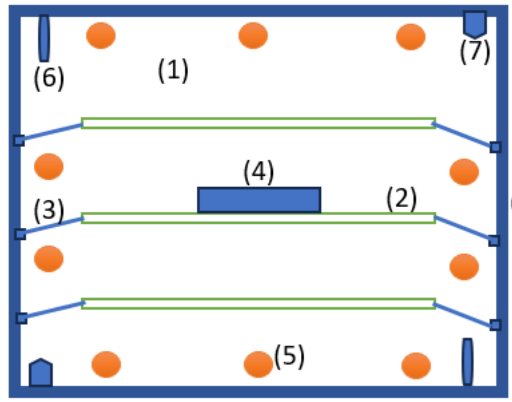

Figure 1b. Experimental freeze-drying chamber

(1)- freeze-drying chamber; (2)- The grid frame for placing the freeze-drying samples, made of 304 stainless steel, is connected to the load cell lever arm; (3)- The lever arm connects the sample grid to the vertically moving slider of the load cell to determine the change in sample mass during the drying process; (4) The yogurt samples, pre-frozen for freeze-drying; (5)- Infrared lamps provide heat to ensure uniform temperature within the drying chamber; (6)- Temperature sensor; (7)- Pressure sensor; (8)- Drying chamber wall with a hole connecting to the freezing chamber and vacuum pump.

The flat slab-shaped yogurt is frozen until the surface temperature reaches -37°C and the core temperature reaches -33°C. At this point, the average temperature of the frozen yogurt slab is approximately -35°C. At this temperature, the water within the frozen yogurt slab is completely solidified, preparing the sample for the subsequent freeze-drying stage, (Hoan Thi Pham, Linh T.K. Do, Tuan Thanh Chau, Dzung Tan Nguyen, 2023).

The initial moisture content of the frozen yogurt flat slab is W_0 (%) and is uniformly distributed throughout the entire sample.

The moisture content of the freeze-dried yogurt product reaches the desired level W_e (%) corresponding to the drying time $t = t_e$ (seconds).

The frozen yogurt flat slab is placed into the freeze-drying chamber with the drying environment set at a temperature T = 30 to 40ºC and pressure P = 0.01 to 0.1 mmHg, which is below 4.58 mmHg. Freeze-drying experiments are conducted under these conditions to investigate and determine the moisture diffusivity coefficient, (Duan, X., Yang, X., Ren, G., Pang, Y., Liu, L., and Liu, Y., 2016; Liviu Giurgiulescu, et al., 2024).

2.2. Development of a Mathematical Model Describing Mass Transfer During Freeze-Drying

Since H, h >> a = 2L (see Figure 1), the Fick's second law model describing the sublimation of frozen water can be expressed as follows (Dzung N.T., Chuyen H.V., Linh V.T.K., et al., 2022):

$$\frac{\partial W}{\partial t} = D\frac{\partial^2 W}{\partial z^2};\ t \geq 0;\ -L \leq z \leq L \qquad (1)$$

- Initial condition:

At t = 0, the moisture content is uniformly distributed:

$$W(z, 0) = W_o = const \qquad (2)$$

- Boundary conditions:

At z = 0 and z = L and at time $t = t_e$:

$$W(0, t_e) = W(L, t_e) = W_e = const \qquad (3)$$

2.3. Solving the Mathematical Model to Determine the Moisture Diffusivity of the Drying Material

Introduce a new function to homogenize the initial and boundary conditions for solving the system of equations (1), (2), and (3), (Nguyen Tan Dzung., 2017):

$$u(z, t) = W(z, t) - W_e \qquad (4)$$

$$\Rightarrow \begin{cases} u(z,0) = W_o - W_e \\ u(0,t) = u(L,t) = 0 \end{cases} \qquad (5)$$

Substituting equation (4) into equation (1) yields:

$$\begin{cases} \dfrac{\partial u(z,t)}{\partial t} = D\dfrac{\partial^2 u(z,t)}{\partial z^2} \\ u(z,t) = u_o = W_o - W_e \\ u(0,t) = 0 \\ u(L,t) = 0 \end{cases} \qquad (6)$$

Let u(z, t) = Z(z).T(t) (7)

Substituting equation (7) into equation (6) yields:

$$\frac{1}{D}\frac{T'(t)}{T(t)} = \frac{Z''(z)}{Z(z)} = -k, (k > 0) \qquad (8)$$

$$(8) \Rightarrow \begin{cases} T'(t) + kD.T(t) = 0 \\ Z''(z) + k\,Z(z) = 0 \end{cases} \qquad (9)$$

Solving system of equations (9) yields the solution:

$$\begin{cases} T(t) = B.\exp(-kDt) \\ Z(z) = C_1\sin\left(z\sqrt{k}\right) + C_2\cos\left(z\sqrt{k}\right) \end{cases} \qquad (10)$$

Substituting equation (10) into equation (7) yields the solution of equation (6):

$$u(z,t) = Z(z).T(t) = \left[C_1\sin\left(z\sqrt{k}\right) + C_2\cos\left(z\sqrt{k}\right)\right]B.\exp(-kDt) \qquad (11)$$

From equation (11), it follows that:

$$u(0,t) = 0 \Rightarrow C_2 = 0 \qquad (12)$$

Substituting equation (12) into equation (11) yields:

$$u(z,t) = \left[C_1\sin\left(z\sqrt{k}\right)\right]B.\exp(-kDt) \qquad (13)$$

From equation (13), it follows that:

$$u(L, t) = 0 \Rightarrow \sin\left(L\sqrt{k}\right) = 0$$

$$\Rightarrow L\sqrt{k} = n\pi \Rightarrow k = \left(\frac{n\pi}{L}\right)^2 \qquad (14)$$

Let $A = B.C_1$, since n = 1 → ∞ the solution (13) can be rewritten in the form of the following series:

$$u(z,t)=\sum_{n=1}^{\infty}A_n\sin\left(\frac{n\pi}{L}z\right).\exp\left[-D.\left(\frac{n\pi}{L}\right)^2 t\right] \quad (15)$$

The coefficients A_n are determined according to the initial condition:

$$u(z,0)=u_o=W_o-W_e=\text{const} \quad (16)$$

From equation (15), it follows that:

$$u_o=\sum_{n=1}^{\infty}A_n\sin\left(\frac{n\pi}{L}z\right)\Leftrightarrow$$

$$2u_o\sum_{n=1}^{\infty}\sin\left(\frac{n\pi}{L}z\right)=\sum_{n=1}^{\infty}2A_n\sin^2\left(\frac{n\pi}{L}z\right)$$

$$\Leftrightarrow A_n\int_0^L 2\sin^2\left(\frac{n\pi}{L}z\right)dz=2u_o\int_0^L\sin\left(\frac{n\pi}{L}z\right)dz$$

$$\Leftrightarrow A_n\int_0^L\left[1-\cos\left(\frac{2n\pi}{L}z\right)\right]dz=$$

$$2u_o\left[-\frac{L}{n\pi}\cos\left(\frac{n\pi}{L}z\right)\right]_0^L$$

$$\Leftrightarrow A_n\left[z-\frac{L}{2n\pi}\sin\left(\frac{2n\pi}{L}z\right)\right]_0^L=$$

$$2u_o\frac{L}{n\pi}\left(1-\cos(n\pi)\right), n=2m+1$$

$$\Leftrightarrow A_n=\frac{4u_o}{n\pi} \quad (17)$$

With: $n=2m+1$

Substituting equation (17) into equation (15) yields the solution:

$$u(z,t)=u_0\sum_{\substack{m=0\\ n=2m+1}}^{\infty}\frac{4}{n\pi}\sin\left(\frac{n\pi}{L}z\right).\exp\left[-D.\left(\frac{n\pi}{L}\right)^2 t\right] \quad (18)$$

Equation (18) is rewritten as follows:

$$W(z,t)=W_e+$$

$$\left(W_o-W_e\right)\sum_{\substack{m=0\\ n=2m+1}}^{\infty}\frac{4}{n\pi}\sin\left(\frac{n\pi}{L}z\right).\exp\left[-D.\left(\frac{n\pi}{L}\right)^2 t\right] \quad (19)$$

Since the moisture content in the drying material is non-uniformly distributed, it is necessary to calculate the average moisture content. The average moisture content of the drying material is determined by the following equation (Nguyen Tan Dzung., 2017):

$$\overline{W}(z,t)=\frac{1}{L}\int_0^L W(z,t)dz=\frac{1}{L}\left[\int_0^L W_e dz+\right.$$

$$\left.\int_0^L\left(W_o-W_e\right)\sum_{\substack{m=0\\ n=2m+1}}^{\infty}\frac{4}{n\pi}\sin\left(\frac{n\pi}{L}z\right).\exp\left[-D.\left(\frac{n\pi}{L}\right)^2 t\right]dz\right] \quad (20)$$

Calculating equation (20) yields:

$$\overline{W}(z,t)=W_e+\frac{\left(W_o-W_e\right)}{L}\times$$

$$\sum_{\substack{m=1\\ n=2m+1}}^{\infty}\frac{4L}{(n\pi)^2}\left[-\cos\left(\frac{n\pi}{L}z\right)\right]_0^L.\exp\left[-D.\left(\frac{n\pi}{L}\right)^2 t\right]$$

$$\Rightarrow\frac{\overline{W}(z,t)-W_e}{W_o-W_e}=\sum_{\substack{m=0\\ n=2m+1}}^{\infty}\frac{8}{(n\pi)^2}.\exp\left[-D.\left(\frac{n\pi}{L}\right)^2.t\right] \quad (21)$$

Since the series (21) converges rapidly when $m \geq 1$, in practice, only the term with $m = 0$ is considered in calculations. Therefore, equation (21) is rewritten as follows:

$$\frac{\overline{W}(z,t)-W_e}{W_o-W_e}=\frac{8}{\pi^2}\times\exp\left[-\frac{\pi^2 D}{L^2}.t\right] \quad (22)$$

$$\text{Let: } MR=\frac{\overline{W}(z,t)-W_e}{W_o-W_e} \quad (23)$$

Rewriting from equations (22) and (23):

$$\ln\left(MR\right)=\ln\left(\frac{8}{\pi^2}\right)-\left(\frac{\pi^2 D}{L^2}\right).t \quad (24)$$

$$\text{Let: } A=\frac{8}{\pi^2}=0.8106;\ b=\frac{\pi^2 D}{L^2} \quad (25)$$

Rewriting from equations (24) and (25):

$$\ln\left(MR\right)=-bt+\ln\left(A\right) \quad (26)$$

Equation (26) serves as the basis for determining the coefficient b, which is then used to experimentally determine the moisture diffusion coefficient of the drying material.

$$D = \frac{L^2 b}{\pi^2}, \ m^2/s \qquad (27)$$

3. Materials and methods

3.1. Raw material

Sample Preparation: The raw material used is yogurt with an initial moisture content W_o = 83% = 0.83. The mold is designed to ensure uniform shape and thickness among samples, facilitating the drying process and subsequent moisture kinetics analysis. The sample is poured into a flat plate mold with the following dimensions (Figure 2):

- Thickness: a = 2L = 4mm = 0.004m; half-thickness: L = 2mm = 0.002m
- Width: h = 30mm = 0.03m
- Length: H = 60mm = 0.06m

Freezing the Sample: The entire mold filled with yogurt is placed in a freezing environment at a temperature of −50°C. The freezing process continues until the surface temperature of the flat plate reaches −37⁰C and the temperature at the center of the flat plate reaches −33⁰C. At this point, the average temperature of the flat yogurt sample is approximately −35⁰C. At this temperature, the water in the sample is completely frozen, ensuring suitable conditions for proceeding to the freeze-drying stage.

Storage of the Frozen Sample: After the freezing process is complete, the flat yogurt sample is removed from the mold and stored in a stable environment at a temperature of −35⁰C. Maintaining this temperature helps preserve the frozen state of the sample, ensuring the necessary initial conditions for the freeze-drying process (Athanasios, I. Liapis and Roberto Bruttini, 2020).

Thus, the sample preparation process has been completed and is ready for the subsequent steps in the study.

Note: The samples were frozen separately to the required temperature before being placed into the freeze-drying chamber. Prior to loading the samples, the chamber was also pre-cooled to approximately the average temperature of the samples. This procedure helps minimize experimental errors.

Moisture Content Requirement of the Freeze-Dried Product: For freeze-dried yogurt products, the technical requirement is that the final moisture content must not exceed 3%. This moisture level is considered the necessary equilibrium moisture content to ensure long-term preservation, inhibit microbial growth, and maintain the sensory and nutritional properties of the product. Therefore, in this study, the post-drying moisture content is selected as W_e = 3% = 0.03 (Yildiz, F., 2010).

3.2. Equipment

The main equipment used in this study is the DS-12 Freeze Drying System, which was designed, and fabricated by the research team of Associate Professor Dr. Dzung N.T from the Department of Chemical and Food Technology, Ho Chi Minh City University of Technology and Education, Vietnam (Dzung, N.T and et al., 2024).

Figure 2. The DS-12 Freeze Drying System automatically measured and controlled by compute

3.3. Methods

3.3.1. Freeze-drying experiment of yogurt

The yogurt samples were frozen and stored at a stable temperature of −35⁰C. These samples were then transferred into the freeze-drying chamber of the DS-12 freeze-dryer system to conduct the experiment.

The drying ambient temperature was set at T = (30 ÷ 40) °C and the drying ambient pressure at P = (0.01 ÷ 0.1) mmHg, which is below 4.58 mmHg, see Table 2 and Table 3. Temperature and pressure were automatically measured and controlled through sensors connected to a computer, with the data displayed on the control panel. The measurement of time was obtained directly from the computer system.

Freeze-drying was carried out to investigate the change in moisture content of yogurt ($\overline{W}(z,t)$) over drying time, t (seconds).

3.3.2. Moisture content of the product

The moisture content of the product ($\overline{W}(z,t)$, %) was determined using the following formula (Dzung, N.T and et al., 2024):

$$\overline{W}(z,t) = 100 - \frac{G_o}{G_j}\left(100 - W_o\right), (\%) \quad (28)$$

Where: G_0 = 10g represents the initial mass of yogurt before drying; G_j (g) represents the mass of yogurt after drying; W_0 represents the initial moisture content of yogurt (%).

In which, W_0 represents the initial moisture content of yogurt (%) was determined using the convective drying method in a drying oven, as described in AOAC – 927.05. Accurately weighed G_0 = 10g of finely ground sample was placed in a clean, dry, pre-weighed aluminum dish. The sample was then subjected to the drying cabinet at a temperature of 105 °C until a constant mass was achieved (AOAC International, 2000);

G_0 = 10g was determined by gravimetric measurement;

G_j (g) was measured using a specially designed mass sensor located inside the drying chamber to monitor the sample's weight changes throughout the drying process, with the data displayed on the control panel.

3.3. Experimental data processing and analysis

For experimental data processing in this study, only Microsoft Excel 2024 combined with statistical theory was used. The Excel Solver function was applied to determine the maximum and minimum values.

In addition, this study employed Matlab 2024 software to simulate the variation of the diffusion coefficient as a function of the temperature and pressure of the freeze-drying environment.

4. Results and Discussion

4.1. Some necessary thermophysical parameters for calculating the moisture diffusion coefficient of yogurt

The frozen yogurt material in flat sheet form has thermophysical properties presented in Table 1.

Table 1. Thermophysical Properties of Yogurt Material

Parameters	Symbol	Unit	Value
Half-thickness	L	m	0.002
Width	b	m	0.03
Length	H	m	0.06
Moisture content of the material	W_0	%	83
Moisture content of the product	W_e	%	3

These thermophysical parameters are essential for calculating the variation of Ln(MR) with drying time *t*, as presented in Equation (26). From this, the coefficient *b* in the linear relationship of Equation (26) can be determined. Subsequently, by applying Equation (27), the effective moisture diffusivity of yogurt during the freeze-drying process can be accurately calculated.

4.2. Determination of the moisture diffusivity of freeze-dried yogurt

The experiment was conducted by freeze-drying yogurt using the DS-12 system at a freeze-drying ambient temperature of T = 30°C and an ambient pressure of P = 0.1 mmHg to investigate the moisture content changes of the material over drying time. Based on this, the moisture ratio (MR) as a function of drying time was calculated. Equation (28) was used to determine $\overline{W}(z,t)$, and equation (23) was applied to calculate the moisture ratio (MR) and Ln(MR). The experimental results and the calculated values are summarized and presented in Table 2.

Table 2. Variation of moisture content ($\overline{W}(z,t)$, %) and moisture ratio (MR) and Ln(MR) over drying time t (s)

t (h)	t (s)	$\overline{W}(z,t)$ %	MR	Ln(MR)-Ln(A)
0	0	0.8300	1.0000	0.2100
1	3600	0.7240	0.8675	0.0678
2	7200	0.6140	0.7300	-0.1047
3	10800	0.4640	0.5425	-0.4016
4	14400	0.3260	0.3700	-0.7843
5	18000	0.2690	0.2988	-0.9982
6	21600	0.2380	0.2600	-1.1371
7	25200	0.2160	0.2325	-1.2489
8	28800	0.1740	0.1800	-1.5048
9	32400	0.1510	0.1513	-1.6788
10	36000	0.1316	0.1270	-1.8536
11	39600	0.1138	0.1048	-2.0462
12	43200	0.1080	0.0975	-2.1179
13	46800	0.0892	0.0740	-2.3937
14	50400	0.0786	0.0608	-2.5910
15	54000	0.0721	0.0526	-2.7346
16	57600	0.0617	0.0396	-3.0183
17	61200	0.0568	0.0335	-3.1862
18	64800	0.0531	0.0289	-3.3348
19	68400	0.0487	0.0234	-3.5461
20	72000	0.0465	0.0206	-3.6713
21	75600	0.0432	0.0164	-3.8982
22	79200	0.0409	0.0136	-4.0859
23	82800	0.0387	0.0109	-4.3113
24	86400	0.0368	0.0085	-4.5577
25	90000	0.0363	0.0078	-4.6420
26	93600	0.0353	0.0066	-4.8126
27	97200	0.0343	0.0053	-5.0254
28	100800	0.0335	0.0044	-5.2219
29	104400	0.0332	0.0040	-5.3115
30	108000	0.0330	0.0038	-5.3760

Based on the data in Table 2 describing the relationship between Ln(MR/A) and t. the least squares method was applied to minimize the sum of squared deviations between experimental values and those calculated from the mathematical model. At the same time, the significance of the model coefficients was tested using the Student's t-test, and the goodness of fit of the mathematical model was evaluated by the Fisher's F-test. The resulting mathematical model was determined as follows:

$$\mathrm{Ln}(MR/A) = -5.5\times10^{-5}\times t \quad (29)$$

With $A = \frac{8}{\pi^2} = 0.8106$, it follows that:

$$\mathrm{Ln}(A) = -0.201$$

Equation (29) is rewritten as follows:

$$\mathrm{Ln}(MR) = -5.5\times10^{-5}\times t - 0.201 \quad (30)$$

The simulation of the relationship between Ln(MR) and drying time (t) using the experimental data in Table 2 and the mathematical model (30) is presented in Figure 3. The correlation coefficient of R^2 = 0.9945 indicates a very strong relationship between Ln(MR) and drying time (t).

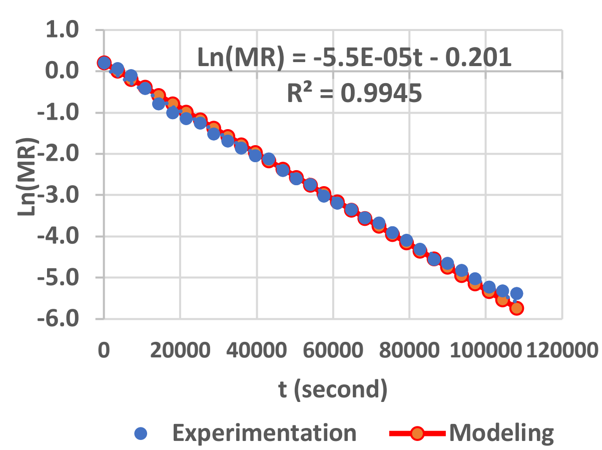

Figure 3. The relationship between Ln(MR) and drying time (t)

From Figure 3, the simulation results as well as the compatibility test of the mathematical model (29) according to the Fisher criterion ($F < F_{1-p}(f_1, f_2)$; $p = 0.05$) demonstrate that the mathematical model (30), which describes the relationship between Ln(MR) and drying time (t), fits the experimental data very well.

Therefore, model (30) can be applied to determine the moisture diffusion coefficient of yogurt during freeze drying. Based on this model, the drying kinetics of yogurt freeze drying can be further analyzed.

From the mathematical model (30), the coefficient b in equation (26) has been determined:

$$Ln(MR) = -5.5\times10^{-5}\times t - 0.201 = -bt - 0.201$$

It follows that: $b = 5.5\times10^{-5}$ (31)

From equations (27) and (31), the following can be determined:

$$D = \frac{L^2 b}{\pi^2} = \frac{0.002^2 \times 5.5\times10^{-5}}{\pi^2} \quad (32)$$

$D = 2.2313\times10^{-11}$ m^2/s

The calculation results showed that, at a sublimation drying environment temperature of $X_1 = T = 30°C$ and a drying environment pressure of $X_2 = P = 0.1$ mmHg, the experiment determined the moisture diffusion coefficient of yogurt during the freeze-drying process to be $D = 2.2313\times10^{-11}$ m^2/s.

Although there is no available data on the moisture diffusion coefficient of yogurt during freeze-drying from other studies for direct comparison, the calculated value is fully consistent with the experimental data and aligns well with previously published research (Athanasios I. Liapis and Roberto Bruttini, 2020; Dzung N. T. (Ed), Chuyen H. Van, Linh V. T. K, Nhiem L.T., Hoan P. T., Linh D.T.K, Quang N.D., Duyen N.D.M, Tung P.T., 2022). Typically, materials subjected to freeze-drying have moisture diffusion coefficients in the range of 10^{-10} to 10^{-12} m^2/s.

The experiment showed that when the temperature and pressure of the freeze-drying environment change, the moisture diffusion coefficient of yogurt during the freeze-drying process also varies accordingly. For details, see the experimental results and calculated values presented in Table 3.

4.3. Build a mathematical model to determine the moisture diffusion coefficient of yogurt during the freeze-drying process

The freeze-drying experiments of yogurt, which were previously frozen during the sample preparation step, were arranged as presented in Table 3. Each experiment used a different set of temperature and pressure parameters for the freeze-drying environment.

The freeze-drying experiments of yogurt were conducted using the sets of temperature and pressure parameters of the drying environment as presented in Table 3. The calculations were performed similarly to those in section 4.2, and the moisture diffusion coefficients of yogurt during the freeze-drying process were determined, compiled, and presented in Table 3.

Table 3. Relationship between the moisture diffusion coefficient of yogurt (D) and the temperature (T) and pressure (P) of the freeze-drying environment

T $T = X_1$ (^oC)	P $P = X_2$ (mmHg)	D (m^2/s) $Y_E = D\times10^{11}$ (m^2/s)
30	0.10	2.231
32	0.08	2.246
34	0.06	2.261
36	0.04	2.273
38	0.02	2.297
40	0.01	2.332
30	0.01	1.830
32	0.02	1.967
34	0.04	2.178
36	0.06	2.422
38	0.08	2.615
40	0.10	2.868

Let $X_1 = T$ (^oC) be the drying environment temperature; $X_2 = P$ (mmHg) be the drying environment pressure; $Y\times10^{11} = D$ (m^2/s) be the moisture diffusion coefficient of yogurt. Therefore, $Y_E = D\times10^{11}$ (m^2/s).

Based on the experimental data in Table 3, the relationship pattern between the moisture diffusion coefficient of yogurt during freeze-drying and the temperature and pressure of the drying environment can be predicted as follows:

$$Y_M = b_o + b_1X_1 + b_2X_2 + b_{12}X_1X_2 \qquad (33)$$

The mathematical model (33) was established based on the experimental data presented in Table 3. Simulation of these data using Matlab 2024 indicated that the relationship between Y_M and X_1, X_2 follows a planar rather than a curved surface. Furthermore, no existing models were adopted in this study, as they are not suitable for the specific case where yogurt serves as the drying material.

From the predictive mathematical model (33) and the experimental data in Table 3, the root means square error (RMSE) function representing the total squared deviation between the calculated values from the model and the experimental data is constructed as follows:

$$RMSE = \sqrt{\frac{1}{N-1}\sum_{j=1}^{N}\left(Y_{Mj} - Y_{Ej}\right)^2} = (b_0, b_1, b_2, b_{12}) \qquad (34)$$

Trong đó: N is the number of experiments (in Table 3, N = 12); j is the index of the j-th experimen; Y_{Mj} is the value calculated from the mathematical model (33) for the j-th experiment; Y_{Ej} is the value determined from the j-th experimental result.

The mathematical model (33) is considered appropriate when the values calculated from the model (Y_M) are equal to or approximately equal to the experimental values (Y_E). Therefore, when the RMSE value reaches its minimum, the model Y_M can be regarded as closely approximating Y_E, from which the model parameters b_0, b_1, b_2, and b_{12} can be determined.

Using Excel – Solver 2024, the following parameters were determined: when $b_0 = 0.284$; $b_1 = 0.05$; $b_2 = 0.123$ and $b_{12} = 0.144$, the minimum value of the Root Mean Square Error (RMSE) was determined to be 0.0124

$$\Leftrightarrow RMSE_{min} = 0.0124$$

The results of the calculations can be seen in Table 4.

Table 4. Calculation of the model values (Equation 33) and the Root Mean Square Error (RMSE)

X_1	X_2	Y_E	Y_M	$(Y_M - Y_E)^2$	δ_E (%)
30	0.10	2.231	2.222	0.0001	0.41
32	0.08	2.246	2.256	0.0001	0.43
34	0.06	2.261	2.278	0.0003	0.75
36	0.04	2.273	2.289	0.0002	0.69
38	0.02	2.297	2.288	0.0001	0.40
40	0.01	2.332	2.334	0.0000	0.07
30	0.01	1.830	1.822	0.0001	0.43
32	0.02	1.967	1.972	0.0000	0.26
34	0.04	2.178	2.177	0.0000	0.01
36	0.06	2.422	2.395	0.0007	**1.11**
38	0.08	2.615	2.624	0.0001	0.33
40	0.10	2.868	2.864	0.0000	0.14
	Σ	**27.519**	**RMSE**	**0.0124**	

Based on the calculation results in Table 4, the minimum value of the RMSE function was determined, from which the mathematical model (Equation 33) was established as follows:

$$Y_M = D\times10^{11} = 0.284 + 0.05\times X_1 + 0.123\times X_2 + 0.144\times X_1\times X_2 \qquad (35)$$

Now, it is necessary to verify the compatibility of the mathematical model (35) with the experimental data. In freeze-drying technology, the maximum allowable relative error of the mathematical model must be less than 5%, that is:

$$\delta_{E\,max} = Max\left\{\frac{|Y_{Mj} - Y_{Ej}|}{Y_{Ej}}\times100\right\} \le 5\% \qquad (36)$$

From Table 4, it is shown that the maximum relative error, $\delta_{Emax} = 1.11$, is less than 5%. Therefore, the mathematical model (35) is compatible with the experimental data. Thus, this model can be used to determine the moisture diffusion coefficient of yogurt during the freeze-drying process, to calculate the drying kinetics, to develop optimal technological conditions, as well as to support the calculation and design of freeze-drying systems.

Once again, to demonstrate the strong relationship between Y_M and X_1 and X_2 in the mathematical model (35), it is necessary to determine the correlation coefficient R^2. The correlation coefficient R^2 is determined as follows:

$$R^2 = 1 - \frac{RSS}{TSS} \qquad (37)$$

In which, N = 12: Number of experiments;

$RSS = \sum_{i=1}^{N}\left(Y_{Ei} - Y_{Mi}\right) =$ 0.0017: Residual Sum of Squares;

$TSS = \sum_{i=1}^{N}\left(Y_{Ei} - \overline{Y}\right) =$ 0.7944: Total Sum of Squares;

$\overline{Y} = \frac{1}{N}\sum_{i=1}^{N} Y_{Ei} = 2.293$: average value of Y_E;

Therefore, the correlation coefficient R^2 is determined as follows:

$$R^2 = 1 - \frac{RSS}{TSS} = 1 - \frac{0.0017}{0.7944} = 0.9979$$

The correlation coefficient of $R^2 = 0.9979$ indicates a very strong relationship between Y_M and X_1 and X_2.

According to the study by Suk Shin Kim and Santi R. Bhowmik (1995) on Effective moisture diffusivity of plain yogurt undergoing microwave vacuum drying, the diffusivity coefficient was found to be primarily influenced by microwave power; R. Lemus-Mondaca et al. (2007). Dehydration characteristics of papaya (Carica pubenscens): determination of diffusion coefficient. The results of the study established the relationship between the effective moisture diffusivity and the drying air temperature; W.J.N. Fernando, H.C. Low, A.L. Ahmad, (2011). The study was conducted on slices of banana, cassava, and pumpkin to determine the dependence of the effective moisture diffusivity on thickness and temperature during convective drying of sliced materials.

The results of the above studies have shown that the effective moisture diffusivity of drying materials depends on many factors and usually ranges from 10^{-12} to 10^{-8} m²/s, which is consistent with the findings of this publication. However, none of the previous studies have established a relationship between moisture diffusivity and the temperature and pressure of the freeze-drying environment for yogurt. Therefore, the development of the mathematical model (37), which describes the relationship between moisture diffusivity and the temperature and pressure of the freeze-drying environment for yogurt, is a completely novel contribution. It plays a significant role in addressing the problem of simultaneous heat and mass transfer in freeze-drying yogurt, thereby determining the drying kinetics and optimizing the technological drying conditions.

The simulation of mathematical model (35) on a 3D coordinate system is presented in Figure 4.

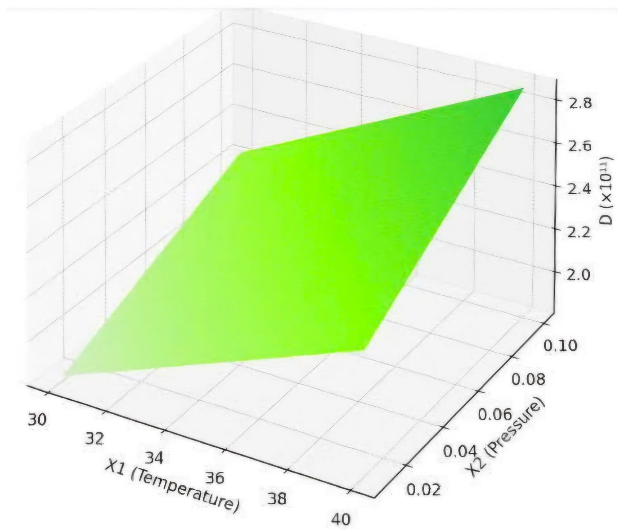

Figure 4. Relationship between the moisture diffusion coefficient of yogurt and the temperature and pressure of the freeze-drying environment.

Looking at the 3D graph, it is evident that the relationship between the temperature and pressure of the freeze-drying environment and the moisture diffusion coefficient of yogurt is linear. This can be considered a novel finding that has not been reported in previous studies. This linear relationship facilitates solving coupled heat and mass transfer problems using numerical methods such as the finite difference method and the finite element method.

From equation (35), it can be rewritten as:

$$D = (0.284 + 0.05\times X_1 + 0.123\times X_2 + 0.144\times X_1\times X_2)\times 10^{-11},\ m^2/s \qquad (38)$$

Where:

$30\ ^oC \le X_1 = T \le 40\ ^oC$

$0.01\ mmHg \le X_2 = P \le 0.1\ mmHg$

The method for constructing the mathematical model (38) to determine the moisture diffusion coefficient of yogurt during freeze-drying, although applied to a specific material—yogurt—essentially generalizes into a universal approach. This is because the method can be applied to any drying material to determine its corresponding moisture diffusion coefficient. This is the core value of this research.

5. Conclusions

This study has proposed a method to determine the moisture diffusion coefficient specifically for yogurt and, more generally, for other drying materials during the freeze-drying process.

Based on this method, the moisture diffusion coefficient of yogurt during the freeze-drying process has been determined through the mathematical model (38). This is an important issue that must be resolved prior to modeling and optimizing the freeze-drying process in order to establish an appropriate technological regime.

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Conflict of interest

The author declares no conflict of interest.

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