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STUDY DYNAMICS OF THE FREEZE DRYING PROCESS OF ROYAL JELLY IN VIET NAM

Nguyen Tan Dzung*

**Department of Food Technology, Faculty of Chemical and Food Technology, HCMC University of Technology and Education, No 01-Vo Van Ngan Street, Thu Duc District, Viet Nam*
tandzung072@yahoo.com.vn

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ABSTRACT

Study dynamics of the freeze drying process of royal jelly in Viet Nam was building and solving mathematical models of heat and mass transfer during the freeze drying process in order to determine relationship between the residual water content of royal jelly product and time of the freeze drying process, $W_M = f(\tau)$. Results obtained were use to set up the technological mode of the freeze drying process of royal jelly in Viet Nam and kinetic parameters of the freeze drying process in order to use to calculate, design and manufacture the freeze drying system in Viet Nam. In addition, they were able to use to set up the technological mode of the freeze drying process for some foods that have high value as royal jelly in Viet Nam.

1. Introduction

The freeze drying is a dehydrate process inside drying material such as food product, pharmaceutical product and types of probiotics. This process is carried out in condition of low temperature and low pressure, under three phase point of water, O(0.0098^{0}C, 4.58mmHg). It can be exactly said that temperature of drying material is lower than the crystallization point of water inside drying material; pressure of drying chamber is lower than 4.58mmHg (Hammami, 1997). As a result, final products after freeze drying have very good quality as the same initial material. For example, protein of final product is not denaturated and be able to recover the original its quality; glucides are not hydrolysed; lipid is not oxidised; ours natural pigments, smells and flavours are not destroyed; bioactive compound and enzymes are not lost activities. In other words, the nutritional value, pharmaceutical value and cosmetics value of product is approximately constant. This is strong point of the freeze drying that have not any drying methods are able to create product as them. However, the freeze drying is quite complicatedly process. At the time, it is not only to happend the heat transfer but also to happend the mass transfer. For this reason, building and solving the mathematical model of heat and mass transfer during freeze drying process were one of difficult problems to answer. Up to now, there were many research on the mathematical models about the heat and mass transfer during freeze drying process to describe the different physical models of drying materials such as flat – shaped model (Luikov, 1975), cylindrical model (Luikov et al., 1975) and spherical model (Luikov et al., 1975), Luikov equation applicable to sublimation – drying (Peng, 1994). Besides, there were also many authors that studied many the mathematical models to describe the complicated physical models such as Heat and mass transfer models for freeze drying of vegetable slices (George and Datta, 2002); Kinetic model for freeze drying process (Boss et al., 2004); Freeze drying pharmaceutical in vials on trays (Liapis et al., 2005); Exergy analysis of freeze drying pharmaceutical in vials on trays (Liapis and Bruttini, 2008), … etc. However, there were not any mathematical model of freeze drying process that was able to apply for the freeze drying of royal jelly; error between experimental data with calculating data of mathematical model is over 36.09%. Therefore, the aim of this study was building and solving heat and mass transfer models during the freeze drying process of royal jelly with parameters of optimal technological mode. Results obtained were used to determine moisture diffusion coefficient, kinetic parameters and calculate, design and manufacture the freeze drying system, establish the technological mode of the freeze drying process for some kind of high food.

2.Building and solving heat and mass transfer models

2. 2.1.Hypotheses building mathematical models

- Royal jelly was frozen in trays, trays are made by glass, trays are rectangular parallelepiped that have area of f (m^2), length of a’ (m), width of b’ (m) and height of δ (m), with a’, b’ >> δ. Therefore, it was supposed that royal jelly in trays is infinite flat – shaped models. The heat and mass transfer during freeze drying process are only conformable to direction height (Oz), i.e. $\partial t/\partial x = \partial t/\partial y = 0$ (Figure 1) (Boss et al., 2004).

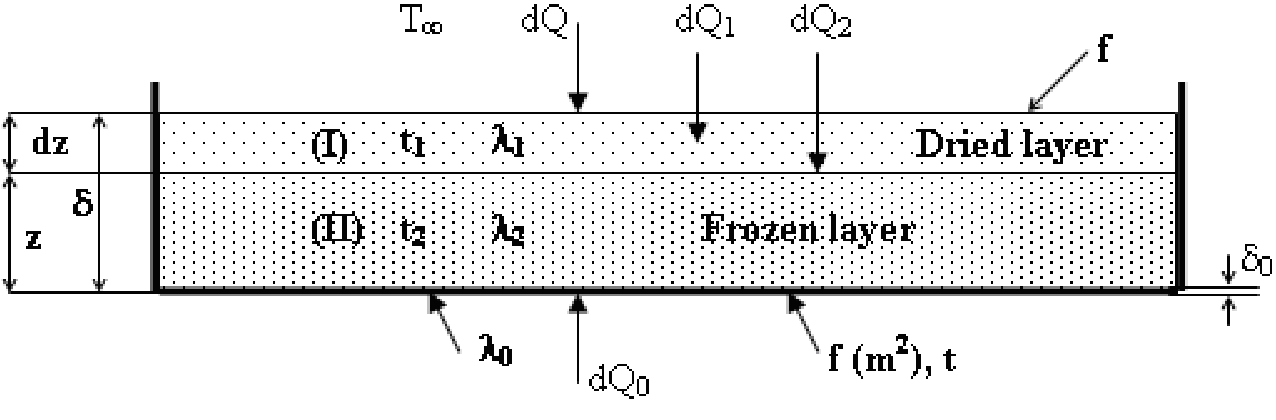

Figure 1. Trays content of royal jelly

- Thermophysical parameters of royal jelly such as initial residual water content of royal jelly W_0; specific heat c_1 (J/(kg.K)), density ρ, ρ_1 (kg/m^3); effective thermal conductivity λ_0, λ_1 (W/(m.K)); thermal diffusivity coefficient a_1 (m^2/s); heat transfer coefficient α_r (W/(m^2.K)); latent heat of sublimation r_{th} (J/kg); latent heat of evaporation r_{hh} (J/kg) were determined, (Liapis and Bruttini, 2008).

2.2.Building the mathematical models for freeze drying of royal jelly

2.2.1.Energy balance equation during the freeze drying of royal jelly

Energy supplied for the freeze drying of royal jelly is energy total of two parts: the first part is energy in order to sublimate ice in area (I); the second part is energy transmitted through area (I) in order to exchange heat with area (II). It can be illustrated as Figure 1

(George and Datta, 2002; Liapis and Bruttini, 2008).

$$dQ + dQ_0 = dQ_1 + dQ_2 \qquad (1)$$

- $dQ + dQ_0$ (J): energy supplied for the freeze drying process of royal jelly.

$$dQ + dQ_0 = -\lambda_1\left(\frac{\partial t_1}{\partial z}\right).f.d\tau - \lambda_0\left(\frac{\partial t}{\partial z}\right).f.d\tau \qquad (2)$$

- dQ_1 (J): energy in order to sublimate ice inside royal jelly in area (I).

$$dQ_1 = r_{th}.\frac{\partial G_w}{\partial \tau}d\tau \qquad (3)$$

- dQ_2 (J): energy transmitted through area (I) in order to exchange heat with area (II).

$$dQ_2 = -\lambda_2\left(\frac{\partial t_2}{\partial z}\right).f.d\tau \qquad (4)$$

Therefore:

$$-\lambda_1\left(\frac{\partial t_1}{\partial z}\right).f.d\tau - \lambda_0\left(\frac{\partial t}{\partial z}\right).f.d\tau = r_{th}.\frac{\partial G_w}{\partial \tau}d\tau - \lambda_2\left(\frac{\partial t_2}{\partial z}\right).f.d\tau \qquad (5)$$

With $t_2 = T_{th} = const$ (Liapis and Bruttini, 2008), thus: $-\lambda_2\left(\frac{\partial t_2}{\partial z}\right) = 0$ (6)

Combination (5) and (6), result obtained was able to written as follow:

$$-\lambda_1\left(\frac{\partial t_1}{\partial z}\right).f - \lambda_0\left(\frac{\partial t}{\partial z}\right).f = r_{th}.\frac{\partial G_w}{\partial \tau} \qquad (7)$$

2.2.2. Heat transfer equation of Fourier

When ice inside royal jelly is not sublimated, whole royal jelly in trays is frozen layer (II), its temperature is constant ($t_2 = T_{th}$ = const). But ice inside royal jelly begins sublimation, surface layer of royal jelly in trays will be sublimated, it created two layers: dried layer (I) and frozen layer (II) is illustrated as Figure 1. Temperature of frozen layer (II) is still constant, temperature of dried layer (I) is root of Equation (8) as follow:

$$\frac{\partial t_1}{\partial \tau} = a_1\frac{\partial^2 t_1}{\partial z^2} \qquad (8)$$

Initial condition:

$$\tau = 0;\ t_0 = t_2 = T_{th} = const. \qquad (9)$$

Temperature of drying chamber is:

$$T_\infty = Z_1 = const; \qquad (10)$$

Boundary conditions:

$$\begin{cases} -\lambda_1\left(\frac{\partial t_1}{\partial z}\right)_{z=\delta} = \alpha_{bx}\left(t_1 - T_\infty\right) \\ -\lambda_1\left(\frac{\partial t_1}{\partial z}\right)_{z=0} = 0 \end{cases} \qquad (11)$$

Solving equation system from Eq. (8) to Eq. (11) was found root as follow:

$$t_1 = T_\infty + \left(T_{th} - T_\infty\right). \to \to \sum_{n=1}^{\infty} A_n.\cos\left(\mu_n.\frac{z}{\delta}\right).\exp\left(-\mu_n^2.Fo\right) \qquad (12)$$

where: μ_n are roots of specific equation:

$$\cot g\mu_n = \frac{\mu_n}{Bi} \qquad (13)$$

Bi is Biot number of dried layer (I):

$$Bi = \frac{\alpha_r.\delta}{\lambda_1} \qquad (14)$$

Fo is Fourier number of dried layer (I):

$$Fo = \frac{a_1\tau}{\delta^2} \qquad (15)$$

A_n is parameters root:

$$A_n = \frac{2\sin\mu_n}{\mu_n + \sin\mu_n \cos\mu_n} \qquad (16)$$

with: $$q_0 = -\lambda_0\left(\frac{\partial t}{\partial z}\right) = \frac{\lambda_0}{\delta_0}\left(T_\infty - T_{th}\right) \qquad (17)$$

$$\alpha_r = \frac{k.C_0.\varepsilon_{qd}}{T_\infty - T_{th}} \int_{T_{th}}^{T_\infty} \left(T^2 + T_{th}^2\right).\left(T + T_{th}\right) dT \qquad (18)$$

with: C_0 = 5.67×10^{-8} W/(m^2.K^4) – thermal radiation coefficient of blackbody (Wei et al., 2005); ε_{qd} – conversion coefficient of dry material that they are transferred by radiation heat, (Holman, 1986); ε_1, ε_2: black level of dry material that that they are transferred by radiation heat, (Gebhart, 1992).

$$\varepsilon_{qd} = \frac{1}{\dfrac{1}{\varepsilon_1} + \dfrac{1}{\varepsilon_2} - 1} \qquad (19)$$

2.2.3. Mass transfer equation of Fick 2

The mass transfer model for the freeze drying process of royal jelly can be written by second law of Fick (Boss et al., 2004) as follow:

$$\frac{\partial G_w}{\partial \tau} = D\frac{\partial^2 G_w}{\partial z^2} \qquad (20)$$

where: D (m^2/s): moisture diffusion coefficient of royal jelly; $G_w = (G.W_0 - G.W)$ is ice mass inside royal jelly that sublimated, W is the residual water content of the royal jelly freeze drying. From Eq. (7) and Eq. (20), it can be written as follow:

$$-\lambda_1\left(\frac{\partial t_1}{\partial z}\right).f - \lambda_0\left(\frac{\partial t}{\partial z}\right).f = r_{th}.D.\frac{\partial^2 G_w}{\partial z^2} =$$
$$r_{th}.D.\frac{\partial^2\left(G.W_0 - G.W\right)}{\partial z^2} = -r_{th}.D.\frac{\partial^2\left(f.\delta.\rho.W\right)}{\partial z^2}$$

As a result:

$$-\lambda_1\left(\frac{\partial t_1}{\partial z}\right).f - \lambda_0\left(\frac{\partial t}{\partial z}\right).f$$
$$= -r_{th}.D.f.\delta.\rho.\frac{\partial^2 W}{\partial z^2} \qquad (21)$$

Corresponding to (21):

$$\frac{\partial^2 W}{\partial z^2} = \frac{\lambda_1}{r_{th}.D.\delta.\rho}.\left(\frac{\partial t_1}{\partial z}\right) - \frac{\lambda_0\left(T_\infty - T_{th}\right)}{r_{th}.D.\delta.\rho.\delta_0} \qquad (22)$$

Substituting Eq. (12) into Eq. (22), result received Eq. (23) as follow:

$$\frac{\partial^2 W}{\partial z^2} = -\frac{\lambda_1}{r_{th}.D.\rho.\delta}.\left(T_{th} - T_\infty\right)\sum_{n=1}^{\infty} A_n.\frac{\mu_n}{\delta}.\rightarrow$$
$$\rightarrow \sin\left(\mu_n \frac{z}{\delta}\right).\exp\left(-\mu_n^2.Fo\right) - \frac{\lambda_0\left(T_\infty - T_{th}\right)}{r_{th}.D.\delta.\rho.\delta_0} \qquad (23)$$

Integrate Eq. (23) found out Eq. (24) as follow:

$$W(z,\tau) = \frac{\lambda_1.\left(T_{th} - T_\infty\right)}{r_{th}.D.\rho}.\sum_{n=1}^{\infty}\frac{A_n}{\mu_n}.\sin\left(\mu_n \frac{z}{\delta}\right).\rightarrow$$
$$\rightarrow \exp\left(-\mu_n^2.Fo\right) - \frac{\lambda_0\left(T_\infty - T_{th}\right)}{2.r_{th}.D.\delta.\rho.\delta_0}x^2 + C_1 z + C_2 \qquad (24)$$

When $\tau = \tau_t$ and z = 0, W = 0 (the residual water content of the Final royal jelly freeze drying calculated by theory), substitute all these value into Eq. (24), result found $C_2 = 0$. For this reason, Eq. (24) can be written as follow:

$$W(z,\tau) = \frac{\lambda_1.\left(T_{th} - T_\infty\right)}{r_{th}.D.\rho}.\sum_{n=1}^{\infty}\frac{A_n}{\mu_n}.\sin\left(\mu_n \frac{z}{\delta}\right).\rightarrow$$
$$\rightarrow \exp\left(-\mu_n^2.Fo\right) - \frac{\lambda_0\left(T_\infty - T_{th}\right)}{2.r_{th}.D.\delta.\rho.\delta_0}z^2 + C_1 z \qquad (25)$$

When τ = 0 and z = δ, W = W_0 (Initial the residual water content of royal jelly), substituting all these value into Eq. (25), result found C_1 as follow:

$$C_1 = \left(W_0 - \frac{\lambda_1.\left(T_{th}-T_\infty\right)}{r_{th}.D.\rho}.\sum_{n=1}^{\infty}\frac{A_n}{\mu_n}.\sin\mu_n\right)/\delta + \frac{\lambda_0.\left(T_\infty - T_{th}\right)}{2.r_{th}.D.\rho.\delta_0} \qquad (26)$$

Substituting Eq. (26) into Eq. (25), result received as follow:

$$W(z,\tau) = \frac{\lambda_1.\left(T_{th}-T_\infty\right)}{r_{th}.D.\rho}.\sum_{n=1}^{\infty}\frac{A_n}{\mu_n}.\sin\left(\mu_n\frac{z}{\delta}\right).\rightarrow$$
$$\rightarrow \exp\left(-\mu_n^2.Fo\right) - \frac{\lambda_0\left(T_\infty - T_{th}\right)}{2.r_{th}.D.\delta.\rho.\delta_0}\left(z^2 - \delta.z\right) +$$
$$\left(W_0 - \frac{\lambda_1.\left(T_{th}-T_\infty\right)}{r_{th}.D.\rho}.\sum_{n=1}^{\infty}\frac{A_n}{\mu_n}.\sin\mu_n\right).\frac{z}{\delta} \qquad (27)$$

2.2.4. Kinetic equation for the freeze drying of royal jelly

The residual water content of royal jelly varying time of the freeze drying process is always determined according to average value of water in volume unit of royal jelly.

$$W_M(\tau) = \frac{1}{\delta}.\int_0^\delta \left(\begin{array}{l} \frac{\lambda_1.\left(T_{th}-T_\infty\right)}{r_{th}.D.\rho}.\sum_{n=1}^{\infty}\frac{A_n}{\mu_n}.\rightarrow \\ \rightarrow \sin\left(\mu_n\frac{z}{\delta}\right).\exp\left(-\mu_n^2.Fo\right) \\ -\frac{\lambda_0\left(T_\infty - T_{th}\right)}{2.r_{th}.D.\delta.\rho.\delta_0}z^2 + C_1 z \end{array}\right) dz$$

(28)

From Eq. (28), kinetic equation for the freeze drying of royal jelly can be written:

$$W_M(\tau) = \frac{\lambda_1\left(T_\infty - T_{th}\right)}{r_{th}.D.\rho}.\sum_{n=1}^{\infty}\frac{2Bi}{\mu_n^2.\left(\mu_n^2 + Bi + Bi^2\right)}.\rightarrow$$
$$\rightarrow \exp\left(-\mu_n^2.Fo\right) + \frac{1}{2}.\left(W_0 + \frac{\lambda_1.\left(T_\infty - T_{th}\right)}{r_{th}.D.\rho}.\rightarrow\right.$$
$$\left.\rightarrow \sum_{n=1}^{\infty}\frac{A_n}{\mu_n}.\sin\mu_n\right) + \frac{\lambda_0.\delta.\left(T_\infty - T_{th}\right)}{12.r_{th}.D.\rho.\delta_0} \qquad (29)$$

3. Materials and methods

3.1. Materials

The royal jelly is harvested from bees's nest to grow up at Bao Loc area in Lam Dong province of Viet Nam. It is the pure natural product and does not mix any chemical composition. It is very thick solution (Sabatini et al, 2009). The basic composition of royal jelly is presented in Table 1 (Dzung, 2013). Before the freeze drying, royal jelly is frozen at the optimal freezing temperature in order that water in royal jelly is completely crystallized. According to research result of Dzung (2014), it was obvious that when royal jelly is frozen until temperature of royal jelly reach $T_{Fopt} = T_{th} = -18.33^0C$. At the time, water inside royal jelly was completely crystallized ω = 1 or ω = 100% (Dzung, 2014).

Table 1. The compositions of royal jelly in Viet Nam

No	Substance	Symbol	Value (% of material weight)	Value (% of dry weight)
1	Water	X_1	59.20	-
2	Proteins	X_2	14.26	34.95
3	Glucids	X_3	15.59	39.09
4	Lipids	X_4	4.00	9.80
5	Minerals	X_5	1.10	2.70
6	10-HDA & impurities	X_6	5.49	13.46

3.2. Apparatus

Equipments used in this study are listed as follow (Dzung, 2013 & 2014):

Figure 2. The freeze drying system DS-3 with the auto-freezing (-50 ÷ - 45)^{0}C

- Determining weigh of royal jelly by Satoriusbasic Type BA310S: range scale (0 ÷ 350)g, error: ± 0.1g = ± 0.0001 kg.
- Determining temperature of royal jelly Dual Digital Thermometer: range scale (-50 ÷ 70)^{0}C, error ± 0.05^{0}C.
- The Freeze Drying System DS-3 (Figure 2) that was controlled automatically by computer. It could reduce the temperature of freezing environment to (-50 ÷ -45)^{0}C. The temperature, pressure and time profile of freeze drying process are measured by computer.

3.3. Methods

- The residual water content of royal jelly of the freeze drying process (W_E, %) is determined by the mass sensor controlled via computer, (Dzung, 2013).

$$W_E = 100 - \frac{G_0}{G_e}\left(100 - W_0\right) \qquad (30)$$

- Using the mathematical tools and computer to solve the mathematical models of heat and mass transfer during freeze drying process of royal jelly (Dzung, 2014).

4. Results and discussions

4. 4.1.Determining moisture diffusion coefficient of royal jelly in freeze drying process

The material properties used for freeze drying of royal jelly in Viet Nam in Table 2 are essential parameters for calculating and simulation Eq. (29), (Figura et al., 2007; Wytrychowski et al., 2013).

Table 2. Thermophysical parameters used for freeze drying of royal jelly in Viet Nam

Symbol	Value	References
W_0 (%)	59.2	Dzung, 2014
δ (m)	12.93×10^{-3}	Dzung, 2014
δ_0 (m)	3.0×10^{-3}	Dzung, 2014
T_{kt} (^{0}C)	-1.06	Dzung, 2014
T_{th} (^{0}C)	-18.33	Dzung, 2014
T_∞ (^{0}C)	20.58	Dzung, 2014

r_{th} (J/kg)	3231.78×10^{3}	Holman, 1986
r_{hh} (J/kg)	2555.65×10^{3}	Holman, 1986
C_0 W/(m^2.K^4)	5.67	Holman, 1986
ε_1	0.96	Heldman, 1992
ε_2	0.91	Heldman, 1992
ε_{qd}	0.8768	Calculation
k	0.9827	Heldman, 1992
ρ (kg/m^3)	1183.22	Heldman, 1992
ρ_1 (kg/m^3)	1328.07	Heldman, 1992
λ_0 (W/(m.K))	1.0183	Dzung, 2014
λ_1 (W/(m.K))	0.1790	Heldman, 1992
c_1 (J/(kg.K))	1681.577	Dzung, 2014
$A_1 = \lambda_1/(c_1.\ \rho_1)$ (m^2/s)	8.0167×10^{-8}	Calculation
α_r	4.4883	Calculation
$Bi = (\alpha_r.\delta)/\lambda_1$	0.3241	Calculation

Substituting thermophysical parameters in Table 2 into the specific Eq. (13). After that solving Eq. (13) was found roots and was presented in Table 3 as follows:

Table 3. Roots of specific equation (13) are μ_j (j = 1 ÷ 19)

Symbol	μ_1	μ_2	μ_3	μ_4
Value	0.5403	3.2413	6.3343	9.459
Symbol	μ_5	μ_6	μ_7	μ_8
Value	12.5921	15.7286	18.8667	22.0059
Symbol	μ_9	μ_{10}	μ_{11}	μ_{12}
Value	25.1456	28.2858	31.4262	34.5669
Symbol	μ_{13}	μ_{14}	μ_{15}	μ_{16}
Value	37.7077	40.8486	43.9897	47.1308
Symbol	μ_{17}	μ_{18}		
Value	50.2719	53.4131		

Eq. (29) can be described the kinetic for freeze drying of royal jelly, if moisture diffusion coefficient of royal jelly D (m^2/s) was determined. However, this kinetic parameter was not determined by the common method, it was determined by identify parameter of the mathematical model method as follow (Dzung, 2014):

Firstly, building the residual Root Mean Square Error (RMSR): between the residual water content of royal jelly from the experimental data and the mathematical model data. Correspondingly, RMSR was written by Eq. (31), (Dzung, 2014):

$$RMSR = f(D) = \sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left(W_E(\tau_i) - W_M(\tau_i)\right)^2} \qquad (31)$$

where: D (m^2/s): moisture diffusion coefficient of royal jelly is variable;

RMSR = f(D) is function;

$W_E(\tau_i)$: the residual water content of royal jelly at time of τ_i from experiment in Table 4;

$W_M(\tau_i)$: the residual water content of royal jelly at time of τ_i from the mathematical model (29);

i = 1 ÷ N: the number of experiments;

τ_i: the time of the freeze drying of royal jelly in Table 4;

This much, the problem finding parameter D of mathematical model (29) was expressed as follows: Finding the root $D^{opt} = D$ in order to objective function of RMSR = f(D) reached the minimum value (Dzung, 2014):

$$RMSR_{min} = Min\{f(D)\} = Min\left\{\sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left(W_E(\tau_i)-W_M(\tau_i)\right)^2}\right\} \quad (32)$$

Secondly, finding the minimum value of RMSR by the meshing method: Setting up value of moisture diffusion coefficient of freeze drying of royal jelly varies in range of $D^{min} = D_0 = 10^{-10}$ to $D^{max} = D_n = 10^{-7}$, with jump is $\Delta D = 0.001\times10^{-10}$ and Step number jump determined is $n = (D^{max} - D^{min})/\Delta D$.

Setting up value of drying time varies in range of $\tau_0 = \tau_{min} = 0h$ to $\tau_n = \tau_{max} = 24h$, with jump is $\Delta\tau = 0.5$ and Step number jump determined is $m = (\tau_{max} - \tau_{min})/\Delta\tau$.

Finally, for $j_1 = 1$ to n, with $D = D_0 + j_1.\Delta D$; for $j_2 = 1$ to m, with $\tau = \tau_0 + j_2.\Delta\tau$. After that calculating $W_M(\tau_{j1})$ by Eq. (29) and RMSR by Eq. (32). A as result, the minimum value of RMSR could be determined by the meshing algorithm on above (Dzung, 2014) programmed in MatLab R2008a software. After solving and calculating, the root of Eq. (32) found the results:

$RMSR_{min} = 0.07676$

When: $D^{opt} = D = 9.50 \times 10^{-9}$ m²/s

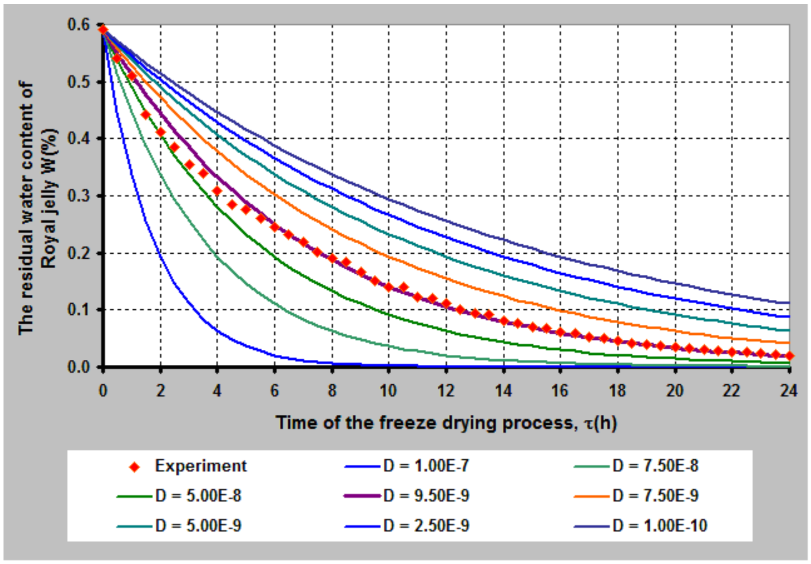

Fig 4. The relationship between residual water content of royal jelly with time of the freeze drying process

From Figure 4, it was obvious that moisture diffusion coefficient of royal jelly was determined by identify parameter of the mathematical model method that was quite complicated and difficult. Because it combined experimental data with mathematical model for freeze drying process of royal jelly. Result on above found out value of moisture diffusion coefficient of royal jelly of $D^{opt} = D = 9.50\times10^{-9}$ m²/s was suitable to describe kinetic as well as calculate and set up technological mode for freeze drying process of royal jelly, (Akay and et al., 1997).

4.2. Test compatibility of the mathematical model

Substituting moisture diffusion coefficient of royal jelly $D = 9.50\times10^{-9}$ m²/s, thermo-physical parameters in Table 2 and roots of specific equation (13) into Eq. (29) with drying time varying from $\tau_0 = 0$ to $\tau_n = 24h$, jump $\Delta\tau = 0.5h$. Results received were shown in Table 4a & b, (Akay et al., 1997; Ratti et al., 2004).

Table 4a. Experiment data and calculating data of the freeze drying of royal jelly

Time (h)	Fo	$W_E(\tau)$	$W_M(\tau)$
0.00	0.0000	0.5920	0.5920
0.50	0.8631	0.5412	0.5499
1.00	1.7262	0.5105	0.5116
1.50	2.5894	0.4419	0.4761
2.00	3.4525	0.4127	0.4430
2.50	4.3156	0.3848	0.4122
3.00	5.1787	0.3548	0.3836
3.50	6.0419	0.3389	0.3569
4.00	6.9050	0.3092	0.3321
4.50	7.7681	0.2845	0.3091
5.00	8.6312	0.2765	0.2876
5.50	9.4944	0.2612	0.2676
6.00	10.3575	0.2456	0.2490
6.50	11.2206	0.2314	0.2317
7.00	12.0837	0.2187	0.2156
7.50	12.9469	0.2013	0.2007
8.00	13.8100	0.1909	0.1867
8.50	14.6731	0.1842	0.1738
9.00	15.5362	0.1667	0.1617
9.50	16.3994	0.1502	0.1505
10.00	17.2625	0.1407	0.1400
10.50	18.1256	0.1392	0.1303
11.00	18.9887	0.1235	0.1212
11.50	19.8518	0.1205	0.1128
12.00	20.7150	0.1107	0.1050
12.50	21.5781	0.1003	0.0977
13.00	22.4412	0.0952	0.0909
13.50	23.3043	0.0914	0.0846
14.00	24.1675	0.0804	0.0787
14.50	25.0306	0.0756	0.0732
15.00	25.8937	0.0701	0.0682
15.50	26.7568	0.0673	0.0634
16.00	27.6200	0.0606	0.0590
16.50	28.4831	0.0593	0.0549
17.00	29.3462	0.0534	0.0511
17.50	30.2093	0.0494	0.0476
18.00	31.0725	0.0457	0.0442
18.50	31.9356	0.0408	0.0412
19.00	32.7987	0.0393	0.0383
19.50	33.6618	0.0379	0.0357
20.00	34.5250	0.0357	0.0332
20.50	35.3881	0.0332	0.0309
21.00	36.2512	0.0312	0.0287
21.50	37.1143	0.0291	0.0267
22.00	37.9774	0.0273	0.0249
22.50	38.8406	0.0252	0.0231
23.00	39.7037	0.0232	0.0215
23.50	40.5668	0.0221	0.0200
24.00	41.4299	0.0201	0.0187

Table 4b. Experiment data and calculating data of the freeze drying of royal jelly

$(W_E - W_M)^2$	$\lvert W_E - W_M \rvert$	Error (%)
1.2326E-32	1.1102E-16	0.00
7.50175E-05	0.00866127	1.60
1.20636E-06	0.00109834	0.22
0.001166728	0.0341574	7.73
0.000917481	0.03028995	7.34
0.000751842	0.02741974	7.13
0.000828683	0.02878686	8.11
0.000325544	0.01804283	5.32
0.000526679	0.0229495	7.42
0.000604094	0.02457831	8.64
0.000123425	0.01110966	4.02
4.13736E-05	0.00643223	2.46
1.18503E-05	0.00344243	1.40
1.18259E-07	0.00034389	0.15
9.32139E-06	0.0030531	1.40
3.99398E-07	0.00063198	0.31
1.73925E-05	0.00417043	2.18
0.000109008	0.01044071	5.67
2.51008E-05	0.00501007	3.01
6.70361E-08	0.00025891	0.17
4.78861E-07	0.000692	0.49
7.95126E-05	0.00891699	6.41
5.1369E-06	0.00226647	1.84
5.90959E-05	0.00768738	6.38
3.2757E-05	0.00572337	5.17
6.83861E-06	0.00261507	2.61
1.84925E-05	0.00430029	4.52
4.64334E-05	0.00681421	7.46
2.8546E-06	0.00168955	2.10
5.55451E-06	0.0023568	3.12
3.78026E-06	0.00194429	2.77
1.5042E-05	0.0038784	5.76
2.50804E-06	0.00158368	2.61
1.92104E-05	0.00438297	7.39
5.27859E-06	0.00229752	4.30
3.41181E-06	0.00184711	3.74

2.10291E-06	0.00145014	3.17
1.41566E-07	0.00037625	0.92
9.6798E-07	0.00098386	2.50
5.04141E-06	0.00224531	5.92
6.35993E-06	0.00252189	7.06
5.41237E-06	0.00232645	7.01
6.10551E-06	0.00247093	7.92
5.60014E-06	0.00236646	8.13
5.87277E-06	0.00242338	8.88
4.20789E-06	0.00205132	8.14
2.75304E-06	0.00165923	7.15
4.2249E-06	0.00205546	9.30
2.09599E-06	0.00144775	7.20

Test compatibility of the mathematical model (29) compared with experimental data is error of the mathematical model (29) with experimental data. It was examined by the Eq. (33) (Gebhart, 1992; Dzung, 2014):

$$\mathrm{Er} = \frac{\left|W_E(\tau_i) - W_M(\tau_i)\right|}{\left|W_E(\tau_i)\right|} \qquad (33)$$

The maximum error of the mathematical model (29) is determined by Eq. (34)

$$\begin{aligned}\mathrm{Er}_{\max} &= \frac{\max\left\{\left|W_E(\tau_i) - W_M(\tau_i)\right|\right\}}{\left|W_E(\tau_i)\right|} \\ &= \frac{\left|0.0221 - 0.02004\right|}{0.0221}.100\% = 9.30\%\end{aligned} \qquad (34)$$

It was able to see that the mathematical model (29) was completely compatible with experimental results. Because of the maximum error of mathematical model (29) with experimental data $\mathrm{Er}_{\max}$ = 9.30%, it is smaller than 10% (Heldman et al., 1992; Dzung, 2014). Therefore, the mathematical model (29) can completely use of describing kinetic for the freeze drying of royal jelly (Holman, 1986).

4.3.Describing kinetic for freeze drying process of royal jelly

From the data in Table 4, it can simulate relationship between residual water content of royal jelly with time of the freeze drying process, $W_E = f(\tau)$ and $W_M = f(\tau)$, (Ratti and et al., 2004). In other words, it can describe kinetic for the freeze drying process of royal jelly. Results obtained were presented in Figure 5. From Figure 5 it is obvious that calculation data from mathematical model (29) are completely suitable with experimental data for the freeze drying process of royal jelly. The average error of the mathematical model (29) compared with experimental data is lower than 9.30%. Therefore, the mathematical model (29) can be used to describe kinetic, calculate and set up technological mode for freeze drying process of royal jelly, (Barresi et al., 2008). The error of mathematical model (29) compared with experimental data was content of all causes as follow: Firstly, mathematical model (29) was calculated by thermophysical parameters of royal jelly of average value and constant, but in the fact that this parameters varied from temperature of royal jelly (Wytrychowski et al., 2013). Secondly, it was supposed that the sublimate temperature of ice inside royal jelly was constant ($t_2 = T_{th}$ = const) to apply for building and solving mathematical model (29), but in the fact that it changed according to time of the freeze drying process, (Barresi et al., 2008). In addition, it was also supposed that physical model of royal jelly in trays was infinite flat – shaped models, but in the fact that it was only approximate as infinite flat – shaped models. Finally, water inside royal jelly had to not be purity water that it was a solution of chemical compositions inside royal jelly and created chemical bonds, physicochemical bonds between water with chemical compositions. For this reason, its latent heat of sublimation and latent heat of evaporation was different compared with purity water (Wytrychowski et al., 2013). All the causes on above made error of mathematical model (29).

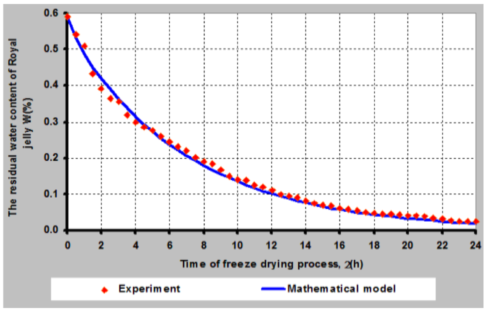

Fig 5. The kinetic for freeze drying process of royal jelly

5. Conclusions

Results of this study were obvious that moisture diffusion coefficient of royal jelly $D^{opt} = D = 9.50\times10^{-9}$ m^2/s was determined by identify parameter of the mathematical model method, this thermophysical parameter was important to calculate physical chemistry processes of the freeze drying (Marine Wytrychowski and et al., 2013). The above results could be concluded that the mathematical model (29) were built by heat and mass transfer models for the freeze drying process of royal jelly. It had well described relationships between the residual water content of royal jelly with time of the freeze drying process. In other words, it had well described kinetic for the freeze drying process of royal jelly. Because it was completely suitable with experimental data, the maximum error of the mathematical model (29) compared with experimental data was 9.30%, it was lower than 10%. Therefore, the mathematical model (29) was used to set up the technological mode of the freeze drying process of royal jelly (Boss et al., 2004; Barresi et al., 2008).

Nomenclature

W_0 (%): initial residual water content of royal jelly; W_E (%): residual water content of royal jelly to determine from experiment; W_M (%): residual water content of royal jelly to determine from mathematical model; G_0 (kg): weight of the initial material royal jelly used for freeze drying; G_e (kg): weight of the royal jelly freeze drying; X_j (%) (j = 1 ÷ 6): the chemical compositions of royal jelly such as water, porteins, glucids, lipids, minerals and impurities.

$c_1 = \sum_{j=2}^{6} c_j X_j = c_{pro}.X_2 + c_{glu}.X_3 + c_{lip}.X_4 + c_{ash}.X_5 + c_{im}.X_6$ (J/(kg.K)): specific heat of dried layer of royal jelly (Heldman, 1992); $c_w = 4167.2 - 9086.4\times10^{-5}\times T + 5473.1\times10^{-6}\times T^2$ (J/(kg.K)), when $0^0C < T < 150^0C$: specific heat of water (Heldman et al., 1992); $c_w = 4080.7 - 5306.2\times10^{-3}\times T + 9951.6\times10^{-4}\times T^2$ (J/(kg.K)), when $-40^0C < T < 0^0C$: specific heat of water (Heldman et al., 1992); $c_i = 2062.3 + 6076.9\times10^{-3}\times T$ (J/(kg.K): specific heat of ice (Heldman, 1992); $c_{pro} = 2008.2 + 1208.9\times10^{-3}\times T + 1312.9\times10^{-6}\times T^2$ (J/(kg.K)): specific heat of protein, (Heldman, 1992); $c_{lip} = 1984.2 + 1473.3\times10^{-3}\times T + 4800.8\times10^{-6}\times T^2$ (J/(kg.K)): specific heat of lipid, (Heldman, 1992); $c_{glu} = 1548.8 + 1962.5\times10^{-3}\times T + 5939.9\times10^{-6}\times T^2$ (J/(kg.K)): specific heat of glucid, (Heldman, 1992); $c_{ash} = 1092.6 + 1889.6\times10^{-3}\times T + 3681.7\times10^{-6}\times T^2$ (J/(kg.K)): specific heat of ash, (Heldman, 1992); $c_{mi} = 1296.78$ (J/(kg.K)): specific heat of impurities inside royal jelly, (Heldman, 1992); $\rho = 1/\sum_{j=1}^{6}\left(X_j/\rho_j\right) = 1/(X_1/\rho_n + X_2/\rho_{pro} + X_3/\rho_{glu} + X_4/\rho_{lip} + X_5/\rho_{ash} + X_6/\rho_{im})$ (kg/m^3): density of royal jelly (Heldman, 1992); $\rho_1 = 1/\sum_{j=2}^{5}\left(X_j/\rho_j\right) = 1/(X_2/\rho_{pro} + X_3/\rho_{glu} + X_4/\rho_{lip} + X_5/\rho_{ash} + X_6/\rho_{im})$ (kg/m^3): density of dried layer of royal jelly (Heldman, 1992); $\rho_w = 1001.75 - 0.4375\times T$ (kg/m^3), with $3.986^0C \leq T \leq 100^0C$: density of water, (Heldman, 1992); $\rho_i = 917\times(1 - 1.55\times10^{-4}\times T)$ (kg/m^3), with $T \leq 0^0C$: density of ice, (Heldman, 1992); $\rho_{pro} = 1329.9 - 0.5184\times T$ (kg/m^3): density of protein (Heldman, 1992); $\rho_{glu} = 1599.1 - 0.31046\times T$ (kg/m^3): density of glucid (Heldman, 1992); $\rho_{lip} = 925.59 - 0.41757\times T$ (kg/m^3): density of lipid (Heldman, 1992); $\rho_{ash} = 2423.8 - 0.28063\times T$ (kg/m^3): density of mineral (Heldman, 1992); $\rho_{im} = 1017.29$ (kg/m^3): density of impurities

(Heldman, 1992); λ_0 (W/(m.K)): effective thermal conductivity of glass (trays), (Heldman, 1992); $\lambda_1 = \sum_{j=2}^{6}\lambda_j x_j$ (W/(m.K)), with $x_j = \left(X_j/\rho_j\right)/\sum_{j=2}^{5}\left(X_j/\rho_j\right)$: effective thermal conductivity of dried layer (Heldman, 1992); $\lambda_{pro} = 0.17881 + 1.1958\times10^{-3}\times T - 2.7178\times10^{-6}\times T^2$ (W/(m.K)): effective thermal conductivity of proteins (Heldman, 1992); $\lambda_{lip} = 0.18071 - 2.7604\times10^{-3}\times T - 1.7749\times10^{-7}\times T^2$ (W/(m.K)): effective thermal conductivity of lipids (Heldman, 1992);

$\lambda_{glu} = 0.20141 + 1.3874\times10^{-3}\times T - 4.3312\times10^{-6}\times T^2$ (W/(m.K)): effective thermal conductivity of glucids (Heldman, 1992); $\lambda_{ash} = 1092.6 + 1889.6\times10^{-3}\times T + 3681.7\times10^{-6}\times T^2$ (W/(m.K)): effective thermal conductivity of minerals (Heldman, 1992); $\lambda_{im} = 1296.78$ (Heldman, 1992); $C_0 = 5.67\times10^{-8}$ W/(m^2.K^4): thermal radiation coefficient of backbody, (Holman, 1986); $\varepsilon_{qd} = 1/(1/\varepsilon_1 + 1/\varepsilon_2 - 1)$: conversion coefficient of dry material that they are transferred by radiation heat (Holman, 1986); ε_1, ε_2: black level of dry material that that they are transferred by radiation heat, (Holman, 1986); D (m^2/s): moisture diffusion coefficeint of royal jelly; $a_1 = \lambda_1/(c_1\times\rho_1)$ (m^2/s): thermal diffusivity coefficient of dried layer (Holman, 1986); α_r (W/(m^2.K): heat transfer coefficient.

$r_{th} = (0.0024\times T^2 + 3.0606\times T + 3287.074)\times10^3$ (J/kg): latent heat of sublimation (Holman, 1986); $r_{hh} = (2509.64 - 2.51\times T)\times10^3$ (J/kg): latent heat of evaporation (Holman, 1986); T_{th} (^{0}C): temperature of ice sublimation; T_∞ (^{0}C): temperature of freeze drying chamber; t_1 (^{0}C): temperature of dried layer; t_2 (^{0}C): temperature of frozen layer; τ (s): time of freeze drying process; f (m^2): heat exchange area of trays; a', b' (m): length and width of trays; $Z_3 = \delta$ (m): height of royal jelly content of trays; δ_0 (m): depth of trays.

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