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Drying kinetics and mathematical modeling of dried macaroni supplemented with Gac aril

Cinética de secado y modelado matemático de macarrones secos suplementados con Gac aril

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Abstract

Macaroni is dried after being boiled to maintain their quality and extend their shelf life. Evaluation of moisture and nutrient changes aims to provide an overview of the phenomena occurring during the drying process. Thin layer drying kinetics of macaroni supplemented with Gac aril in hot air drying was investigated for temperatures range of 60 to 90ºC with six theoretical models were evaluated. It was observed that the Logarithmic equation gave the best prediction to the drying kinetics evidenced by high coefficient of determination (R^2 ranging from 0.98-0.99). The lowest RMSE (0.0103 to 0.0426) and χ^2 values (0.0002 to 0.0024) were obtained from the Logarithmic model within the defined temperature range. Diffusivity coefficients of moisture transfer were found ranging from 1.64×10^{-12} to 2.31×10^{-12} m^2s^{-1} with activation energy was determined at 11.81 $kJ.mol^{-1}$. Drying at 80ºC for 9 hours was selected for this product, the degradation kinetics of β-carotene and lycopene in macaroni followed a first-order kinetic model with half-life values were determined.
Keywords: drying, Gac aril, macaroni, modeling, bioactive compounds.

Resumen

Los macarrones se secan después de hervirlos para mantener su calidad y prolongar su vida útil. La evaluación de los cambios de humedad y nutrientes tiene como objetivo proporcionar una visión general de los fenómenos que ocurren durante el proceso de secado. Se investigó la cinética de secado en capa fina de macarrones suplementados con Gac aril en secado con aire caliente para un rango de temperaturas de 60 a 90 ºC con seis modelos teóricos. Se observó que la ecuación logarítmica dio la mejor predicción de la cinética de secado evidenciada por un alto coeficiente de determinación (R^2 que oscila entre 0,98 y 0,99). Los valores más bajos de RMSE (0,0103 a 0,0426) y χ^2 (0,0002 a 0,0024) se obtuvieron del modelo logarítmico dentro del rango de temperatura definido. Se encontraron coeficientes de difusividad de transferencia de humedad que oscilaron entre $1{,}64 \times10^{-12}$ y $2{,}31\times10^{-12}$ m^2s^{-1} y la energía de activación se determinó en 11,81 kJ.mol-1. Para este producto se seleccionó el secado a 80ºC durante 9 horas, se determinó la cinética de degradación del β-caroteno y licopeno en macarrones siguiendo un modelo cinético de primer orden con valores de vida media.
Palabras clave: secado, gac aril, macarrones, modelado, compuestos bioactivos.

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1 Introduction

The major ingredient of macaroni is wheat flour, which is then blended with other common ingredients. In recent years, it has become more and more common to consume foods that have been nutrient- and color-fortified. In order to increase product quality by using natural colours, adding Gac aril throughout the macaroni-processing process can be a smart idea. According to Thavamany *et al.* (2020), gac includes priceless bioactive components with significant anti-inflammatory, anti-ulcer, anti-ulcer ageing, and antibacterial properties. As the report of Thavamany *et al.* (2020), Gac aril contained up to 500 mg/100 g of lycopence and 73.7 mg/100 g of total phenolic content. A large amount of carotenoids (β-carotene, lycopene), α-tocopherol along with fatty acids present in Gac membranes (Ishida *et al.*, 2004) have made this material widely used in medicine and human nutrition. In addition, the research by Bruno *et al.* (2018) revealed that compared to tomato and watermelon, which contained 2.5% and 20.7% of total fatty acids, respectively, Gac fruit had a greater concentration of oleic acid, accounting for 44.5% of total fatty acids.

After production, macaroni can be boiled and used immediately, the product is only stored for a short time. Drying is one of the common food production method, which improves food stability and increases shelf life. In fact, drying also reduces the weight of the product, simplifies transportation and reduces storage space. Drying can prevent the growth of microorganisms that cause food spoilage and minimizes the physico-chemical changes of food during storage (Thuy *et al.*, 2021). Mathematical modeling is also a frequently used to study food drying kinetics, improving existing drying systems or for new designs for efficient control of the drying process (Doymaz, 2017). Drying process involves simultaneous mass and heat transmission in an unstable condition, which is the most crucial step in the production of pasta. Two parallel mechanisms, involving the transfer of heat from moving air to pasta and the transfer of mass from pasta to moving air, take place when the damp pasta is forced against the drying airflow (Espinosa-Solares & Domínguez-Puerto, 2023; Luna-Flores *et al.*, 2023). In order to optimise the drying process and raise the quality of pasta, it is important to have a proper understanding the heat and mass transfer in pasta. There has been extensive research into the relationship between pasta characteristics and drying conditions. The quality of cooked pasta is influenced by the drying temperature, and drying at a temperature between 60 and 90°C is reported to create pasta of a high quality properties (Lin *et al.*, 2023; Ceccanti *et al.*, 2022) Along with the development of the manufacturing process, development of the model is also taking place. Therefore, it is also important to choose an appropriate model for a particular manufacturing process. Besides that, although hot air is the traditional drying method in drying food, it can however damage product quality, controlling moisture and changing bioactive compounds/nutrients in products is a matter of concern. According to several research, drying techniques have an impact on the physicochemical properties, bioactive components, and antioxidant capacity of foods (Kittibunchakul *et al.*, 2022; Kumar *et al.*, 2022; Mohammadi *et al.*, 2020). However, it has limited documents to present the drying techniques affect the bioactive substances in macaroni that are thought to have the purported health benefits. The purpose of this study is to present and analyze the characteristics related to the macaroni drying model, focusing on the theoretical model, and to propose an appropriate mathematical model describing the characteristics of Macaroni thin-layer drying with the addition of Gac aril. The first-order kinetic model was used to evaluate the degradation kinetics of the beta-carotene and lycopene in macaroni during drying.

2 Material and methods

2.1 Gac puree preparation

In a natural environment, gac fruit (*Momordica cochinchinensis*) is grown at Can Tho University (Can Tho, Vietnam). When the gac fruit was fully red, it was picked (Thuy and Tuyen, 2013). The aril that surround the seeds of the Gac fruit are first removed, followed by a preliminary wash and halving. Gac arils were puree, frozen at -18ºC, and used in the study.

2.2 Macaroni processing

Prepare a mixture of 300 gr (239 g wheat flour, 6 g semolina and 55 g potato starch). Then add the other ingredients that have been fixed weighing (mass enough for a batch of macaroni made in an automatic machine), including 65 g eggs, 63 g water and 2 g salt and the proportions of Gac aril of 7% and xanthan gum 1.5% were calculated according to the total weight of the mixture.

The PHILIPS Macaroni Maker (Japan) should be filled with the flour mixture. Set a 5-minute timer to mix the ingredients. Incubate the dough for 30 minutes while continuing to thoroughly mix it for 5 minutes (Thuy *et al.*, 2020). When enough time is up, start the machine to press the macaroni into a infinite slab (0.4 cm of diameter and 35 cm of length). Macaroni was pre-gelatinized at boiling temperature for 5 minutes and then the quality was analyzed.

Table 1. Predictive mathematical models for food product drying.

Model name	Equation
Henderson and Pabis	MR=a.e^{-kt}
Modified Henderson and Pabis	MR = a.e^{-kt}+b.e^{-gt}+c.e^{-ht}
Logarithmic	MR=a.e^{-kt}+c
Newton	MR= e^{-kt}
Page	MR=e^{-ktn}
Two-term exponential	MR= a.e^{-kt}+(1-a)e^{-kat}

Where t is drying time (hrs); a, b, c, h, n, k are the model constants.

2.3 *Macaroni drying*

2.3.1 *Drying procedure*

The drying process of cooked macaroni was conducted in an oven dryer (Memmert UN30, Germany) at 0.5 m/s of air velocity. The sample was put in a single layer on the stainless-steel tray. During the process, the weight loss was periodically recorded with the accuracy of ±0.01g. The drying process of macaroni was stopped when its moisture content (MC) reached approximately 10%. All drying experiments were performed in triplicate.

2.3.2 *Mathematical modeling*

In this study, we considered samples having geometry in the form of an infinite slab because the macaroni was as above-mentioned. Six mathematical models that are frequently used to describe the drying behaviour were fitted with the drying curves that were acquired from the experiments, which presented in Table 1 (Thuy *et al*., 2021; Tai *et al*., 2021; Loan *et al*., 2023). These were modified based on information from the drying of boiled macaroni.

The ideal mathematical model was chosen by converting the moisture content at different temperatures to moisture ratio (MR), which presents the dimensionless MR (Eq. 1).

$$MR = \frac{M_t - M_e}{M_0 - M_e} \tag{1}$$

where M_o is the product's starting MC, M_e is the equilibrium MC, and M_t is the product's instantaneous MC (kg water/kg dry matter).

In regression analysis, the most suitable model for macaroni drying at various temperatures was determined using statistical calculations involving the correlation coefficient (R^2), chi-square (χ^2), and mean error square average (RMSE). The lowest values of χ^2 and RMSE, along with the highest R^2 value, show that the experimental data and the mathematical model agree, according to Tai *et al*. (2021) and Loan *et al*. (2023).

2.3.3 *Calculation of the activation energy and effective moisture diffusivity*

Due to the infinite slab shape of sample, Eq. 2 described by Crank (1979), which can be applied in the situation of drying of macaroni, assuming one-way moisture flow, negligible shrinkage, constant diffusivity, and initially uneven moisture distribution (Thorat *et al*., 2012).

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

where L is the plate's half thickness (in metres), D_{eff} is the effective diffusivity (measured in m^2/s), t is the drying time (measured in seconds), and n is a positive integer. According to Doymaz (2017), the natural logarithm of both sides is used to create Eq. 3.

$$\ln(MR) = \ln\left(\frac{8}{\pi^2}\right) - \frac{\pi^2}{4L^2}D_{eff}t \tag{3}$$

D_{eff} may be calculated from Eq. 4 (Zarein *et al*., 2015).

$$\text{Slope} = \frac{\pi^2 D_{eff}}{4L^2} \tag{4}$$

The activation energy (E_a) iscalculated through an Arrhenius-type equation (Eq. 5) (Sanjuán *et al*., 2003).

$$D_{eff} = D_0 \exp\left(-\frac{Ea}{RT}\right) \tag{5}$$

where R is the universal gas constant (8,314 J/mol K), T is the absolute drying air temperature (K), D_o is the diffusion coefficient corresponding to infinite temperature (m^2/s), Ea is the activation energy (kJ/mol), and R is the universal gas constant.

2.3.4 *Chemical compositions analysis and sensory evaluation*

Moisture content of product was determined by AOAC (2005). The β-carotene and lycopene content were determined according to the method of Fikselová *et al*. (2008) and Kakubari *et al*. (2020), respectively. The sensory evaluation was conducted to selected highest acceptance sensory properties by various temperature. The sensory evaluation was processed as the description of Thuy *et al*. (2020) and Too *et al*. (2022).

2.3.5 Degradation kinetic

The degradation rate of lycopence and beta-carotence during drying could be described by first-order model (Equation 6) under constant temperature condition:

$$\ln(C/C_0) = -kt \tag{6}$$

where C_o is the initial concentration of bioactive compounds (mg/g), C is bioactive compounds content at the treatment time (mg/g), k is the degradation rate constant (1/hour), and t is the treatment time (hour).

3 Results and discussion

3.1 Effect of drying temperatures

The effect of drying temperature (from 60 to 90ºC) on the drying process of macaroni thin layer was recorded. It was observed that the percentage of moisture decreases continuously with time at different drying temperatures (Figure 1). The final moisture content of macaroni reached about lower 10% (DW) after 12, 11, 9, and 8 hours at drying temperatures of 60, 70, 80 and 90ºC, respectively. It can be seen that as drying time increased, the drying rate also increased and the higher drying rate when the drying air temperature is higher and as a result the drying time required to achieve the final moisture content is shorter.

3.2 Modelling in mathematics

In order to execute curve fitting calculations with drying time using the six drying models, the moisture content data from the various experimental modes were transformed to the more relevant moisture ratio expression. The findings of the statistical analysis

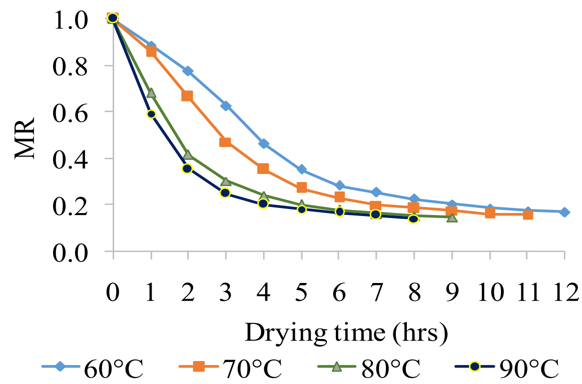

Figure 1. Change of MR (M/M_o) of macaroni with drying time at temperatures from 60 to 90ºC

conducted on these drying model data are presented in Table 2. Based on the Root Mean Square Error (RMSE), coefficient of determination (R^2), and Chi-square (χ^2), the models were assessed. The R^2 values for each equation were consistently high, falling between 93.90 and 99.90%. This showed that all equations were capable of accurately describing the thin layer drying rates of macaroni. RMSE was in the range of 0.013 to 0.075, and 2 was between 0.0002 and 0.0074.

Among six thin layer drying models, the Logarithmic model (2008) shows a higher compatibility (at all drying temperatures performed) than other models with relatively high correlation coefficients ($R^2 \geq 98.25\%$). Similarly, the lowest RMSE (0.0103 to 0.0426) and χ^2 (0.0002 to 0.0024) values were obtained in the Logarithmic model in the defined temperature range. Zhou *et al*. (2015) also reported that the Logarithmic model is more suitable to describe the drying process of non-fried instant noodles than other experimental models at the temperature of 80-120ºC. At temperatures between 60 and 90°C, macaroni can be dried well with the addition of Gac aril. A comparison of models and data was made to assess the goodness-of-fit. The experimentally obtained MR and the predicted MR were found to be satisfactorily correlated in the temperatures investigated, with a high R^2 (0.99).

Table 2. Modelling the drying process of macaroni at different temperatures

Model	Temp (ºC)	Model constants	RSME	R^2 (%)	χ^2
Henderson and Pabis (2015)	60	a=1.0358; k=0.1856	0.0438	97.96	0.0023
	70	a=1.0117; k=0.2278	0.0486	97.50	0.0028
	80	a=0.9966; k=0.3143	0.0506	97.56	0.0032
	90	a=0.9389; k=0.3832	0.0757	93.90	0.0074
Modified Henderson and Pabis (2008)	60	a=0.0062; k=0.2057; b=0.5254; g=0.2061; c=0.5254; h=0.2061	0.0470	98.51	0.0041
	70	a=0.0185; k=0.1598; b=0.5137; g=0.2743; c=0.5137; h=0.2743	0.0399	98.99	0.0032

	80	a=0.1045; k=0.0344; b=0.4522; g=0.5082; c=0.4522; h=0.5082	0.0213	99.75	0.0011
	90	a=0.1485; k=0.0029; b=0.4276; g=0.6847; c=0.4276; h=0.6848	0.0146	99.90	0.0006
Logarithmic (2008)	60	a=0.9869; k=0.2186; c=0.0672	0.0426	98.25	0.0024
	70	a=0.9444; k=0.3048; c=0.1024	0.0369	98.70	0.0018
	80	a=0.8714; k=0.5359; c=0.1391	0.0165	99.73	0.0004
	90	a=0.8580; k=0.6815; c=0.1456	0.0103	99.90	0.0002
Newton (2007)	60	k=0.1784	0.0442	97.74	0.0021
	70	k=0.2248	0.0465	97.48	0.0024
	80	k=0.3411	0.0647	94.76	0.0047
	90	k=0.4132	0.0744	93.26	0.0062
Page (2008)	60	k=0.1606; n=1.0605	0.0472	96.75	0.0027
	70	k=0.2427; n=0.9508	0.048	97.56	0.0028
	80	k=0.4893; n=0.7055	0.0402	98.2	0.002
	90	k=0.6167; n=0.6250	0.0368	98.56	0.0017
Two-term exponential (2007)	60	a=0.4371; k=0.2891	0.0482	97.54	0.0027
	70	a=0.4418; k=0.3621	0.0445	97.9	0.0024
	80	a=0.3903; k=0.5789	0.0432	98.23	0.0023
	90	a=0.2746; k=1.1318	0.0569	96.55	0.0042

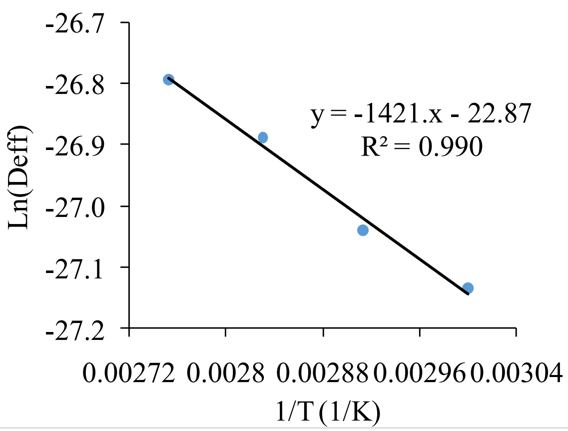

Figure 2. Effective diffusivity and air temperature.

3.3 *The activation energy and moisture diffusivity*

With higher drying temperatures, macaroni's effective moisture dispersion capability significantly increased. At temperatures between 60 and 90°C, dried macaroni's effective diffusivity values ranged from 1.642 $\times 10^{-12}$ to 2.310 $\times 10^{-12}$ m^2/s. The highest D_{eff} values (2.310 $\times 10^{-12}$ m^2/s) were obtained during drying at 90°C. The values were determined to be 4.125. $\times 10^{-11}$ and 8.772 $\times 10^{-11}$ m^2/s, respectively (Goksu *et al.*, 2005), which are lower than the estimated D_{eff} of macaroni beads in the fluidized bed and microwave aided fluidized bed drying. Figure 2 illustrates a plot of the values of ln(D_{eff}) versus 1/T (1/K) to determine how temperature affects the effective diffusivity.

Over the tested temperature range, it was discovered that the graph was linear, demonstrating the Arrhenius dependence. It was determined that the activation energy was 11.81 kJ/mol after calculating it from the slope of the straight line. This value is similar to the pasta drying (11.4 kJ/mol) at 40 to 80ºC (Villeneuve and Gélinas, 2007), but lower than olive cake (12.43 kJ/mol) reported by Vega-Gálvez *et al.* (2010) and dried banana slice (32.65 kJ/mol) from Doymaz (2010).

3.4 *Lycopene and -carotene degradation rates during hot air drying of dried macaroni*

The sensory evaluation was conducted and shown that the macaroni was highest acceptance by 50 panelists when the macaroni supplemented with gac was dried at a drying temperature of 80ºC for 9 hrs. Therefore, the change of lycopene and β-carotene, two main components in Gac aril, were observed. Degradation of lycopene and β-carotene during drying time was recorded. It was observed that the kinetics of lycopene and β-carotene degradation in Gac supplemented macaroni followed a first-order kinetic model, where C is the concentration (μg/g DM) at time t, C_o is the initial concentration of lycopene/β-carotene and k is the degradation rate constant. The initial concentration lycopene and β-carotene of bolied macaroni were 15.26 μg/g and 61.26 μg/g, respectively (Thuy *et al.*, 2023) when applying Gac aril in macaroni. β -carotene helps consumers who lack vitamin A and is good for eye health, and lycopene has been related to a variety of health advantages, including protection against sunburn and some types of cancer as well as improvements in heart health.

Table 3.The rate constant and half-life values of lycopene/β-carotene in Gac supplemented macaroni at drying temperature of 80ºC.

Carotenoids	The rate constant (k)	R^2	$t_{1/2}$ (hours)
Lycopene	0.047±0.003*	0.97	14.8
β-carotene	0.066±0.002	0.99	10.5

*Standard error of regression.

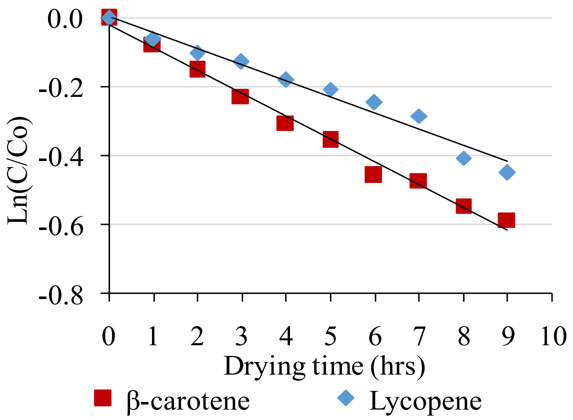

Figure 3. The degradation kinetics of lycopene/β-carotene in macaroni dried at 80°C.

As a result, introducing Gac fruit during the production of macaroni boosted the product's nutritional value and provided users with health benefits. The natural logarithmic plots of lycopene/β-carotene concentrations over time for each temperature are shown in Figure 3.

The correlation coefficient of determination (R^2) between 0.97 and 0.99, was confirmed that the degradation of both components was first-order. The kinetic data for lycopene and β-carotene degraded in macaroni were shown in Table 3. The half life time was calculated as 14.8 hrs for lycopene and 10.5 hrs for β-carotene at 80ºC. It was observed that the higher k value, corresponding to a lower hafl-life value was obtained. In this case, the destruction of lycopene in the product occurs less than that of β-carotene. Baysal *et al*. (2000) reported that the drying process can lead to different degrees of lycopene degradation. Drying tomatoes at 80°C did not cause a significant loss of lycopene content was found by Zanoni *et al*. (1998). Dried macaroni products can be stored at room temperature for more than 3 months.

Conclusions

Changes in the moisture content of macaroni during the drying were noted and successfully predicted by Logarithmic model. With the change in moisture ratio, the decomposition kinetics of lycopene and β-carotene in Gac supplemented macaroni during drying was also evaluated. The estimated $t_{1/2}$ values of lycopence and β-carotene in macaroni was observed at value of 14.8 hours and 10.5 hours, respectively. The results obtained from this study support a better understanding of the effects of drying temperature and time on the healthful ingredients of this product. Knowledge of these effects will be helpful in determining the right conditions for producing macaroni supplemented with Gac fruit of the desired quality.

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