Kinetic Model of Moisture Diffusivity in Soursop Leaves (Annona muricata L.) by Convection Drying

T T Y Nhi1,2,3,*, P V Thinh1,2, N D Vu2,3, N T Bay4, N T M Tho4, N N Quyen5, T T Truc6

1Center of Excellence for Biochemistry and Natural Products, Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam.
2NTT Hi-Tech Institute, Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam.
3Graduate University of Science and Technology, Vietnam Academy of Science and Technology, Ha Noi, Vietnam
4TRAVIPHA Co., Ltd., Tien Giang Province, Vietnam
5Faculty of Environmental and Food Engineering, Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam
6College of Agriculture, Can Tho University, Can Tho City, Vietnam

*Corresponding author: ttynhi@ntt.edu.vn, labasm2013@gmail.com

Abstract. Tea is considered to be a product that brings health benefits. The drying process showed high efficiency in processing this product. However, the process of moisture is not adequately controlled, resulting in wasted energy in the drying process. The dynamics of the drying process of soursop leaves and the effect of temperature on the process speed were assessed in the present study. Five models (Newton, Henderson and Pabis, Page, Logarithmic, and Weibull) were used to determine the suitable drying dynamic at temperatures of 50°C, 55°C, 60°C and 65°C. The mechanism of the processing is based on the principle of dehumidification of the convection drying system with hot air, which caused the moisture inside the material to move outwards. The results showed that the drying temperature is directly proportional to the drying rate of the process, as the increase in drying temperature from 50°C to 65°C produced the link breaking energy and heating that leads to evaporation. Dehumidification rate depends on the temperature, change in Deff value as 1 x (RTa) -1 from from 9,264 95 × 10−9 m/s² to 9.76485 × 10−9 m/s². Page model with determination coefficient R²> 0.99 was found to be the most suitable model for experimental data.

1. Introduction
Drying products are known as diversified and well-preserved food with extended shelf-life [1], [2]. Moreover, foods that contain nutritional compounds are more concerned in the processing industry [3], [4]. In recent years, tea production is increasing rapidly. In particular, tea made from soursop leaves is especially preferred in global market.

Soursop (Annona muricata) belongs to Annonaceae family from the U.S. and Caribbean. [5]–[7]. Soursop leaf has two sides with different colors, the upper part catches the light while the lower part is rough and includes many alkaloids, chlorofill and polyphenol [8]–[10]. Previous studies have illustrated the biological activity of Soursop leaves and fruits. Flavonoids and flavones are the two
biological compounds that are present in high contents in soursop leaves, known to prevent inflammation and cancer in colon epithelia [11]–[13]. The plant leaf extract has been combined with fruit juice to produce jelly candy [2], [14], or encapsulated for preservation purpose by natural mucus[12]. However, studies on the evaporation of tea from soursop leaves are limited. Olalusi et al, 2019 has shown a comparison between the three traditional drying methods on the same material [15]. In addition, a model for Thin Layer Drying Cymbopogon citratus using hot air was developed, which calculated the effective diffusion model and activation energy of the process [16]. With similar attempt, the present study aims to propose five mathematical models of drying dynamics within the temperature range from 50 to 65°C. The linear and non-linear chart were constructed based on the data set on the moisture ratio with the predicted value of R² at a 95% level of confidence. The drying kinetic models provide insight into the moisture escaping from plant materials and understand the mechanism of the process. Besides, the process's kinetic parameters have successfully calculated activation energy, which is necessary for the moisture evacuation process and choosing the right parameters for the drying process to get the most benefit. Therefore, the mathematical techniques used to estimate the kinetics, mechanisms, and energy required are an indispensable step in understanding and practicing experimental drying.

2. Materials and methods

2.1 Materials
A. Muricata leaves from the province of Tien Giang, Vietnam was collected in January. Before drying, the materials were thoroughly washed and cut into uniform size with 17±3 cm of length and 0.54 mm of thickness (Figure 1). The initial moisture content of the leaves was measured as 2.29 (g water/g dry matter) following the method described previously [16].

![Figure 1. Fresh soursop (A. muricata) leave](image)

2.2 Drying Equipment
An oven with hot air flow with maximum temperature of 70°C directly contacted with the material surface. A support blower was employed to blow out moisture at 60Hz frequency. The oven was designed with flexible adjustment and equipped with an auto heat sensor and 12 drying trays (40 × 30 cm) with mesh hole of 1cm in diameter.

![Figure 2. Convection oven (A), and drying tray (B)](image)
2.3 Drying procedure
Leaf samples of *A. muricata* (500g) were spread onto the tray dryer. The temperature for air-drying was selected as 50, 55, 60 and 65°C. The weight for sample was registered every 10 minutes. Experimental data were used to test drying curve mathematical models and measure the efficient diffusivity of humidity and activation energy.

2.4 Determination of mathematical components
The mathematical model for drying kinetics of *A. muricata* leaves was developed based on the moisture ratio, initial drying rated and moisture content. These values were carried out by using the equations as described by Premi *et al.* (2010) [17]. The mathematical models that have been employed to construct the drying kinetics curve and their references were summarized in Table 1. These kinetic models are understood as semi-theoretical and experimental models and are widely applied to many materials. These models provide a higher degree of accuracy and better prediction of the drying process's moisture loss mechanism [18].

Subsequently, they were evaluated for their fitness by applying non-linear regression statistical analysis for coefficient of determination ($R^2$) and chi-square ($\chi^2$), following the method by Togrul (2006) [19]. The fittest model was indicated by having high $R^2$ and the low $\chi^2$ values.

Table 1. Mathematical models for the determination of the drying kinetics of *A. muricata* leaves.

| No | Model name                  | Model                          | References                  |
|----|-----------------------------|-------------------------------|-----------------------------|
| 1  | Newton                      | $MR = \exp(-kt)$ (4)          | Hii and others (2008) [20]  |
| 2  | Henderson and Pabis model   | $MR = a \exp(-kt)$ (5)        | Akoy (2014) [21]            |
| 3  | Page model                  | $MR = \exp(-kt^n)$ (6)        | Hashim and others (2014) [22]|
| 4  | Logarithmic model           | $MR = a \exp(-kt) + c$ (7)    | Rayaguru and Routray (2012) [23]|
| 5  | Weibull                     | $MR = a - b \exp(-kt^n)$ (8)  | Tzempelikos and others (2015) [24]|

Moreover, the activation energy of the drying process and effective moisture diffusion for *A. muricata* leaf samples were determined using the Arrhenius equation and Fick diffusion model, respectively [20].

2.5. Statistical analysis
The moisture ratio, moisture content, efficient diffusivity of the moisture and activation energy of the drying process were calculated Microsoft Excel software (Redmond, WA, USA). The fit mathematical model and their coefficients were determined by Origin Pro software 9.0 (USA).

3. Results and discussion

3.1 Drying Curves and Drying Rate Curves
In this analysis, a drying temperature range of 50-65°C was chosen, which is usually feasible for many medicinal plants [26]. One of the main factors affecting a product's drying kinetics during the air-drying process is known as temperature [27-29]. Results from the present study have shown that increasing temperature would require a shortened drying time (Figure 3). In particular, as the temperature rised from 50°C to 65°C, the drying time was reduced from 80 to 50 min. Such observation could be due to two following reasons: evaporation and moisture diffusion from the materials surface to environment. Similar results were also obtained from another study by Hwa *et al.* (2008) [30].
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The drying rate curves of soursop leaves at different temperatures (50-65°C) were clearly different (Figure 3). High drying temperatures were associated with increased drying rates. This process was performed through (i) initial period accelerating (0-10 minutes) to (iii) falling-rate period (over 30 minutes) while (ii) the constant-rate period (10-30 minute) was ignored because of the thin-layer spread of soursop leaves and high surface moisture removal rate, thus accelerating evaporation and circumventing the saturation state of the material. Studies also have indicated that the constant rate period occurred quite rapidly in most drying processes of fruits and vegetables [18]. In addition, increasing drying temperature has also increased the energy of breaking the bonding, leading to evaporation.

3.2 Mathematical Models of Drying Curves

Tables 1 summarized the regression analyses of the soursop leaves drying process using five semi-theoretical models at different temperatures. The regression parameters, determination coefficient ($R^2$) and chi-square ($\chi^2$) were employed to select the model that best fitted for the experimental data. The model with the highest $R^2$ value and the lowest $\chi^2$ was selected to describe the thin layer drying characteristic. In all cases, the $R^2$ and $\chi^2$ value ranged from 0.91 to 0.9994, and $7.89 \times 10^{-5}$ to 0.05123,
respectively. From the evaluation criteria, all models have well described the drying characteristic of soursop leaves. By using statistical analysis and the estimated coefficients of the mathematical models, the Page model has achieved the highest $R^2$ value (averaged at 0.99885) and the lowest $\chi^2$ value (averaging at 1.37E-4). The result indicated that the Page model is the fittest model to measure the moisture ratio of soursop, followed by the Newton model in which $R^2$, $\chi^2$ averaged at 0.99807 and 2.28 x 10^-4, respectively. These results are similar to the work by Wilton et al. [31], where the Page model was the best to describe the drying characteristics at the temperature range 40°C to 70°C. Similarly, Page model also satisfactorily described the drying behaviour of the slices at the temperature range 50°C to 80°C when using the thin-layer modelling of FHIA-21 (Tetraploid Plantain) using hot-air dryer [32].

Table 2 showed that $R^2$ and $\chi^2$ values of Newton model were weaker than Page model. Meanwhile, the Weibull model exhibited higher $R^2$ and lower $\chi^2$ than other models. Moreover, the Page model estimated that the drying constants $k$ and $n$ increased when increasing the temperature from 50 to 65°C. The drying rate constant value $k$ increased from 0.05835 to 0.15240 at the temperatures of 50°C and 55°C.

**Table 2.** Effects of statistical studies on moisture content modeling and drying time. (min)

| DT  | Drying model | Coefficient | Statistical test |
|-----|--------------|-------------|-----------------|
|     |              | $R^2$       | $\chi^2$ (Chi-square) |
| 50°C | Newton       | k 0.05981   | 0.9982 2.04E-04 |
| 55°C | Newton       | k 0.09027   | 0.9955 5.26E-04 |
| 60°C | Newton       | k 0.10011   | 0.9994 7.44E-05 |
| 65°C | Newton       | k 0.08008   | 0.9992 1.09E-04 |
| 50°C | Page         | k 0.05835 n 1.00800 | 0.9980 2.32E-04 |
| 55°C | Page         | k 0.15240 n 0.81149 | 0.9987 1.51E-04 |
| 60°C | Page         | k 0.11115 n 0.96060 | 0.9994 7.54E-05 |
| 65°C | Page         | k 0.06925 n 1.05143 | 0.9993 9.18E-05 |
| 50°C | Henderson and Pabis | a 0.99942 k 0.05978 | 0.9979 2.33E-04 |
| 55°C | Henderson and Pabis | a 0.99073 k 0.08954 | 0.9948 5.99E-04 |
| 60°C | Henderson and Pabis | a 0.99908 k 0.10004 | 0.9993 8.91E-05 |
| 65°C | Henderson and Pabis | a 1.00225 k 0.08023 | 0.9990 1.30E-04 |
| 50°C | Logarithmic  | a 1.00964 k 0.05729 c -0.01386 | 0.9983 1.96E-04 |
| 55°C | Logarithmic  | a 0.97838 k 0.09445 c 0.01537 | 0.9951 5.64E-04 |
| 60°C | Logarithmic  | a 0.99313 k 0.10226 c 0.00685 | 0.9994 7.89E-05 |
| 65°C | Logarithmic  | a 1.01052 k 0.07788 c -0.00994 | 0.9992 1.13E-04 |
| 50°C | Weibull      | a -0.02546 b -1.02528 k0 0.06759 n 0.93822 | 0.9836 1.86E-04 |
| 55°C | Weibull      | a -0.01568 b -1.01547 k0 0.16963 n 0.75685 | 0.9985 1.77E-04 |
| 60°C | Weibull      | a 0.00662 b -0.99340 k0 0.10295 n 0.99723 | 0.9992 1.05E-04 |
| 65°C | Weibull      | a 0.13168 b -0.86832 k0 0.96215 n 1.03355 | 0.9157 0.05124 |
However, after increasing the drying temperature up to 60°C and 65°C, the drying rate constants tended to decrease to 0.11115 and 0.06925, respectively. Besides, the n values also increased from 0.81149 to 1.05143 when the temperature was between 55°C and 65°C. In term of temperatures, During the entire process, which has also occurred in other agricultural products, drying rates have decreased [33]–[35]. This finding was consistent with previous research, where it was found that the Page model well represents the Plantain Sample drying kinetics. [33].

3.3 Effective moisture diffusivity
The relative linear relationship between drying time (t) and ln(MR) at 50-65°C of drying temperature was shown in Figure 5. The R^2 correlation coefficient and effective moisture diffusion coefficient were calculated by linear regression.

![Figure 5. The change of ln (MR) by time of soursop leaves](image)

| Temperature | Slope  | R^2      | D_{eff} (m/s^2) |
|-------------|--------|----------|-----------------|
| 50°C        | -0.07768 | 0.967228 | 9.26495E-09     |
| 55°C        | -0.07787 | 0.990359 | 9.39551E-09     |
| 60°C        | -0.08093 | 0.969209 | 9.65301E-09     |
| 65°C        | -0.08187 | 0.990593 | 9.76485E-09     |

Table 3 showed that the D_{eff} value increased from 9.26495×10^{-9} m/s^2 to 9.76485×10^{-9} m/s^2, which fell within the normal range of D_{eff} value in typical drying processes for food products [36]. At the temperatures from 50°C to 65°C, the D_{eff} values were between 9.26495×10^{-9} and 9.76485×10^{-9} m/s^2, indicating a proportional rise between the effective diffusivity coefficient and temperature [37]. The moisture content of soursop leaves peaked at 65°C, due to rapid water evaporation from the material surface. Typically, 90% of the energy activation value for vegetables is in the range of 14.42-230.61kJ with a large volume of materials. Activation energy for soursop leave was low (3,357kJ), which had a positive effect on evaporation and power saving. This measurement was also influenced by many values, typically the weight and oven volume [37].
4. Conclusion
The present study has constructed a suitable mathematical model to describe the drying kinetics of soursop leaf, along with the activation energy and the moisture diffusion coefficient of the material. The results showed that high drying temperature was associated with faster moisture removal rate within a shortened period of drying time. The process included the initial and falling-rate periods without the constant-rate period. The Page model was the most appropriate model for explaining hot air thin-layer drying. The diffusion of moisture peaked at 65°C, and the activation energy was relatively high, indicating that more power was needed to remove water from the material by drying.

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