Mathematical modelling of drying kinetics of oven dried 
*Hibiscus Sabdariffa* seed

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**Abstract.** *Hibiscus sabdariffa* or Roselle seed is an agricultural product which possesses a large amount of nutritional content that could be useful in wide application. Drying process can be helpful to conserve the seeds for long storage periods. For this reason, the drying behavior of Roselle seeds was investigated using laboratory oven at various drying temperatures (35, 45, 55, 60 and 65°C). It is found that the drying process of the Roselle seeds consists of constant rate and mostly falling rate period at where the moisture content of Roselle seed decreases as time goes. Adequately suitable drying temperature is found to be 65°C with the drying time of 240 minutes. The drying behavior of Roselle seeds is further understood through the drying kinetics of the Roselle seeds. Five empirical mathematical models were selected to describe and compare the drying characteristics of Roselle seeds at respective drying temperatures. Drying rate constants, coefficients and statistical parameters were determined by non-linear regression analysis, and therefore, the diffusion approach model was selected as the best model with R value obtained is 0.999 while MBE, RMSE and χ² values of 2.37E-04, 0.01335 and 0.007794 respectively.

1. Introduction
Roselle or *Hibiscus sabdariffa Linn* is an ideal crop for ASEAN countries because it is easy to grow and has many uses and application. The plant is a shrub, grown in tropical dry weather and thought of native to Asia or Tropical Africa [1]. It is known to contain proteins, fats, flavonoids, carbohydrates, acids, vitamins minerals and has been reported to have antihypertensive, anticancer, antioxidant, antihyperlipidemic and hepatoprotective properties [2]. Comprises of edible leaves, seeds and calyces has made Roselle used widely in various applications which include dried foods, dessert toppings, juices and syrups, tea, drugs, health supplements, and more. In Malaysia, *Hibiscus sabdariffa Linn* is listed as one of the potential herbs in propelling the sustainability of the nation’s bio-economy sector for the nation’s agriculture National Key Economic Area (NKEA). Along with it, other herbs listed include Tongkat Ali, Kacip Fatimah, Misai Kucing, Hempedu Bumi, Dukung Anak, Mas Cotek, Mengkudu, ginger, Belalai Gajah and Pegaga. The market potential has been studied and the Natural Resources and Environment Ministry recommended the usage of Roselle for medicinal, food, healthcare and beauty.

Drying of agro products is one of the most energy-consuming processes in the food industry, however it is a promising method of reducing postharvest losses. Instead of letting the harvested crops to go rotten...
and decayed, drying can be done to hinder wastage. Throughout the years, many models have been developed in order to model drying curves. By utilising the experimental data, the variables of the models can be determined so that the drying curves can best be represented. In carrying out an effective drying process, the information on the mechanism of moisture removal during the drying process and modelling expressions would be useful for the design and optimisation of the dryers [3]. Effective optimisation of the drying operation can also be contributed by the understanding of the drying process and the raw material characteristics.

Drying process can be described using theoretical and empirical models which are available from a very simple to an extremely complex. Appropriate model is important not just for the type of equipment, but for the level of experimental data available and type of results required. Various drying models is analyzed and selected if it is best fitted for the drying kinetics of drying materials.

The objectives of this work are to investigate the drying characteristics of the Roselle seeds through the effect of drying temperature and time on Roselle seeds experimentally and to develop a drying model for drying kinetics of Roselle seeds and to validate the model by experimental data.

2. Materials and Methods

2.1. Drying experiments
In this research project, drying kinetics of Roselle seeds was investigated as a function of drying conditions with varying temperatures. Drying process of the seeds was done in the laboratory oven using seeds freshly separated from its calyces. Quantity of the seeds used for each experiment run was 10 grams and three readings were taken for each of the run. The drying process was done with temperature ranging from 35 °C to 65 °C. The reading of the seeds weight was obtained every 5 minutes interval for up to several hours until the moisture content was almost constant. Generally, drying temperature of 50 to 60°C is feasible for a wide number of natural product [4]. Low drying temperature of 30-50°C may protect sensitive active ingredient, however it may promote fungi growth due to longer drying time [5]. Meanwhile, higher drying temperature may reduce the product quality due to degradation process.

In studying the effects of the drying on the Roselle seeds, it is necessary to gain knowledge of the drying kinetics because it is considered important for the design, simulation and optimisation of the process using mathematical modelling. This was done to determine the optimum drying temperature and drying time for the drying process or the Roselle seeds. Drying kinetics of the Roselle seeds was evaluated experimentally by obtaining the weight of the drying sample which was the seeds as a function of time [6]. In this study, the drying curves was presented in average moisture content versus time and drying rate versus average moisture content. The percentage of moisture content was obtained using the Equation 1:

\[
\text{Moisture content (\%)} = \frac{w_2 - w_3}{w_2 - w_1} \times 100\% \quad (1)
\]

Where
- \( w_1 \) is initial weight of tray (g)
- \( w_2 \) is weight of tray containing sample before drying (g)
- \( w_3 \) is weight of tray containing sample after drying (g)

2.2 Mathematical modelling of drying curves
Mathematical modelling has importance in the prediction of the drying process of the Roselle seeds. The common models used for drying are Newton, Page, Modified Page and, Henderson and Pabis model. Analysis of data was done by obtaining the rate of drying curves in the constant-drying conditions for each temperature. Moisture ratio (MR) of sample was expressed as \((M - M_2)/(M_0 - M_2)\). The accuracy of the model was indicated by correlation coefficient (R) which was the parameter that showed the goodness of fit of model and cannot be greater than unity. Additional statistical parameters which are chi-squared (\( \chi^2 \)), mean bias error (MBE) and root mean square error (RMSE) are calculated using following equations:
\[ \chi^2 = \frac{\sum(M_{\text{exp},i} - M_{\text{pre},i})^2}{N-n} \] (2)

\[ MBE = \frac{1}{N} \sum(M_{\text{pre},i} - M_{\text{exp},i}) \] (3)

\[ RMSE = \left[ \frac{1}{N} \sum(M_{\text{pre},i} - M_{\text{exp},i})^2 \right]^{1/2} \] (4)

3. Result and Discussion

3.1 Drying characteristics

The measurement of the material moisture content versus time formed the drying curves. Drying curves is produced to determine the effect of temperature on drying behaviour of Roselle using drying experimental data obtained by varying temperatures (35°C - 65°C). Drying kinetics of Roselle mainly depends on the drying temperature [6]. The result of the drying process is represented in Figure 1, showing the drying curves obtained at varying temperatures across drying time. The figure shows the visual judgement of the effects of operating temperature on the drying curves of Roselle seeds, at constant drying conditions.

![Figure 1. Roselle seeds drying curves at varying temperatures (35°C to 65°C)](image)

The changes in experimental moisture ratio as a function of drying time of Roselle seeds at different drying temperatures (35°C to 65°C) shown that the drying time required to reduce the moisture from initial moisture to final moisture content was 560, 390, 300, 260 and 240 minutes at drying temperatures of 35, 45, 55, 60 and 65°C respectively. The drying process is considered completed when the moisture ratio achieved its final value which approximately equals to zero as there is no significant changes in the moisture ratio. The moisture ratio decreased exponentially over time for all temperature in this study, which is similar to the behavior of some biological materials [7].

The drying kinetics in the following Figure 2 shown that the drying of Roselle seeds occur at constant rate period, which the MR ranging from 0.9 to 0.7 and mostly at falling rate period ranging from MR of 0.78 to complete drying for temperature range of 35°C to 65°C, thus suggesting that the surface of drying material is less saturated with water.
The seeds undergone falling rate period and constant rate period throughout the drying process at each drying temperature. The process started with the initial moisture content and decreases gradually as time goes until it reaches a point of constant drying rate until equilibrium moisture content is achieved. These phenomena satisfy Fick’s Law of diffusion where the concentrated particle in a stationary medium diffused in the direction that is proportional to the concentration gradient [8].

During constant drying rate period, the rate of drying are constant at which it can be seen as the straight linear line in the plot of moisture versus time. At this period, diffusion occurs by which moisture is evaporated from a saturated surface through a stationary air film that is in contact with the surface. This period depends on the drying condition which in turn determines the temperature of the saturated surface. During this period, water is transported to the surface at a rate sufficient to maintain saturation [9].

During the falling drying rate period, the rate of drying starts to decrease and eventually falls even more rapidly until the equilibrium moisture content is achieved. At this period, moisture reaches the surface at decreasing rate and since the surface is no longer saturated, it tends to rise above the wet bulb temperature. When the drying rates further decreasing, the movement of moisture occurs by diffusion under the concentration gradient created by the depletion of water at the surface. The capacity of the drying air to absorb moisture determines the drying rate and it also depends on the drying air condition [9].

The variation of drying rate with moisture content and drying time are shown in Figure 2 and Figure 3. Drying rate decreases continuously with time and decreasing moisture content. As indicated in these curves, there are falling and constant drying rate period in the drying of Roselle seeds. This shows physical mechanism governing moisture movement in Roselle seeds is dominant by diffusion [10]. According to mass transfer controlling process, the drying rate is controlled by internal diffusion phenomenon [11].
Figure 3. Drying rate versus drying time of Roselle seeds

The higher the drying temperature, the larger is the difference between the saturated and partial pressure of water vapor in the drying air. Initially at early phase, the rate of drying is rapid due to large amount of water present. As drying proceeds, the water content decreases. At final stages, only small amount of water available and thus the process becomes slow.

The drying temperature of 65°C reflected the best temperature as it dry the Roselle seeds at 240 minutes drying time which is faster compared to other drying temperatures. The seeds have undergone a short constant rate period which is between 5 to 10 minutes of drying time and MR of 0.9 down to 0.78. While the remaining process from 10 minutes until the drying completed at 240 minutes, the Roselle seeds have undergone falling rate period. The MR of the falling rate period dropped from 0.78 to approximately zero when the process completed.

It is obvious that the drying process progress with the increment of the operating drying temperature. Similar behaviour reported by several authors of drying experiments [6,10,12,13]. This shows that higher temperature implies larger driving force for heat transfer, and accelerates the drying of material, as the temperature provides a large water vapor pressure deficit [6].

The slight data fluctuation obtained may be due to several errors and uncontrolled parameters during the drying experiment. The unavoidable equipment error has only been minimised by running a few sets of drying experiments and taking the average data obtained from the drying process. The parameter that had not been controlled during the drying process is size of the seed. Seed sizing directly influenced the surface area involved in the drying process. Low uniformity of the sizing of the seeds may influence the process through the heat transfer from seed surrounding to the seeds interior.

3.2 Mathematical modelling of the drying kinetics

The fitted drying models can be evaluated by different criteria to determine the quality of the models. In the analysis of data, the moisture ratio kinetics is essentials to describe the drying of Roselle seeds which is presented in Figure 1. Statistical computations are done to assess the consistency of models for the data depicted in the drying curves of the seeds. It is done for a few most common drying models, which are Newton model, Page model, Henderson & Pabis model, Diffusion approach and Logarithmic model. The details of the results are presented in Table 1.
| Models          | Constants | R        | $\chi^2$  | RMSE   | MBE    |
|----------------|-----------|----------|-----------|--------|--------|
|                |           |          |           |        |        |
| **35°C**       | Newton    | k: 0.00558 | 0.999584  | 0.000125 | 0.011016 | 0.003354 |
|                | Page      | k: 0.00691 | 0.999564  | 7.03E-05 | 0.008155 | 0.001762 |
|                |           | n: 0.95784 |           |        |        |        |
|                | Henderson & Pabis | a: 0.97267 | 0.99686  | 6.5E-05 | 0.007842 | -0.00111 |
|                |           | k: 0.00536 |           |        |        |        |
|                | Diffusion approach | a: 0.97267 | 0.99804  | 3.28E-05 | 0.005492 | 0.000561 |
|                |           | b: 21.7611 |           |        |        |        |
|                |           | k: 0.00536 |           |        |        |        |
|                | Logarithmic | a: 0.48368 | 0.291844 | 0.071908 | 0.257055 | 2.09E-12 |
|                |           | c: 0.51632 |           |        |        |        |
|                |           | k: 83.8774 |           |        |        |        |
| **45°C**       | Newton    | k: 0.00928 | 0.99238  | 0.000194 | 0.013728 | 0.003666 |
|                | Page      | k: 0.00798 | 0.99264  | 0.000167 | 0.012564 | 0.004027 |
|                |           | n: 1.03203 |           |        |        |        |
|                | Henderson & Pabis | a: 1.00122 | 0.99221  | 0.000199 | 0.013721 | 0.003782 |
|                |           | k: 0.00929 |           |        |        |        |
|                | Diffusion approach | a: 6.98470 | 0.99426  | 0.000134 | 0.01111 | 0.003578 |
|                |           | b: 1.04499 |           |        |        |        |
|                |           | k: 0.01190 |           |        |        |        |
|                | Logarithmic | a: 0.62228 | 0.325626 | 0.093429 | 0.293008 | -1.1E-10 |
|                |           | c: 0.37772 |           |        |        |        |
|                |           | k: 118.922 |           |        |        |        |
| **55°C**       | Newton    | k: 0.01553 | 0.997643 | 0.001132 | 0.033033 | 0.004533 |
|                | Page      | k: 0.00680 | 0.998912 | 0.000287 | 0.016315 | 0.005422 |
|                |           | n: 1.19595 |           |        |        |        |
|                | Henderson & Pabis | a: 1.04487 | 0.996913 | 0.00091 | 0.029075 | 0.008563 |
|                |           | k: 0.01616 |           |        |        |        |
|                | Diffusion approach | a: 22.8340 | 0.99907  | 0.00026 | 0.015222 | 0.012216 |
|                |           | b: 1.02730 |           |        |        |        |
|                |           | k: 0.02586 |           |        |        |        |
|                | Logarithmic | a: 0.65602 | 0.369334 | 0.105092 | 0.306321 | -2.4E-10 |
|                |           | c: 0.34398 |           |        |        |        |
|                |           | k: 207.097 |           |        |        |        |
| **60°C**       | Newton    | k: 0.01867 | 0.996854 | 0.00144 | 0.037205 | 0.004421 |
|                | Page      | k: 0.00715 | 0.998926 | 0.000278 | 0.016006 | 0.005149 |
|                |           | n: 1.24105 |           |        |        |        |
|                | Henderson & Pabis | a: 1.05472 | 0.996009 | 0.001128 | 0.032273 | 0.009103 |
|                |           | k: 0.01985 |           |        |        |        |
|                | Diffusion approach | a: 14.5313 | 0.997844 | 0.000592 | 0.022875 | 0.020088 |
|                |           | b: 0.96167 |           |        |        |        |
|                |           | k: 0.01118 |           |        |        |        |
|                | Logarithmic | a: 0.67762 | 0.398402 | 0.101743 | 0.300005 | 1.36E-10 |
|                |           | c: 0.32238 |           |        |        |        |
|                |           | k: 287.822 |           |        |        |        |
| **65°C**       | Newton    | k: 0.02464 | 0.996901 | 0.001031 | 0.031458 | 0.002413 |
|                | Page      | k: 0.01076 | 0.999211 | 0.000178 | 0.012812 | 0.001622 |
|                |           | n: 1.22041 |           |        |        |        |
|                | Henderson & Pabis | a: 1.04989 | 0.996577 | 0.00081 | 0.0273 | 0.005887 |
|                |           | k: 0.02597 |           |        |        |        |
|                | Diffusion approach | a: 17.1611 | 0.999278 | 0.000165 | 0.012062 | 0.01032 |
|                |           | b: 1.03906 |           |        |        |        |
|                |           | k: 0.04179 |           |        |        |        |
|                | Logarithmic | a: 0.72770 | 0.453447 | 0.089277 | 0.280292 | -5E-10 |
|                |           | c: 0.27229 |           |        |        |        |
|                |           | k: 89.2990 |           |        |        |        |
From the statistical analysis, the value of correlation coefficients (R), mean bias error (MBE), root mean square error (RMSE) and chi-square ($\chi^2$) ranges from 0.2918 to 0.9998, 3.28E-05 to 0.10509, 0.005492 to 0.30632 and -5E-10 to 0.020088 respectively for that particular models. The most suitable model to describe the drying is the model giving highest coefficient of determination, lowest Root Mean Square Error (RMSE) and the lowest chi-square ($\chi^2$). Therefore, the diffusion approach model presented highest values at almost all drying temperature as compared to other tested models. The obtained of R reflected 99% strength of the diffusion approach model data and the experimental data.

Therefore, the most acceptable model is presented as

$$MR = a \exp(-kt) + (1 - a) \exp(-kt).$$

The expression is used to estimate the moisture content of Roselle seeds at any time during the process accurately. The performance of the model is illustrated in Figure 5 below. The experimental data seem closely scattered around the line representing computation data. This indicates the suitability of the model to describe drying behavior of Roselle seeds.

**Figure 4.** Experimental and predicted moisture contents of Roselle seeds

From these results, it can be concluded that the diffusion approach model represent the best as compared to other drying models for the oven drying of Roselle seeds. Besides its simplicity, the experimental data align most to the predicted drying data as indicated in Figure 5. It can be observed that there is a good correlation between experimental and predicted MR data. Diffusion approach model has also been reported by other authors as an adequate model to fit drying kinetics of biological materials experiments [7,14].

4. Conclusion

The drying process of Roselle seeds consists of both constant and falling rate period. The drying time required to remove the moisture content was 560, 390, 300, 260 and 240 minutes at temperatures of 35, 45, 55, 60 and 65°C respectively. It can be concluded that increasing drying temperature causes the drying process to complete more rapidly. Therefore, 65°C is a suitable drying temperature with the drying time of 240 minutes. A suitable mathematical drying model is selected for the similar laboratory oven as the drying equipment, by comparing a few most common drying models. In ensuring effective drying process, the diffusion approach model is found to be an adequately suitable to describe the drying kinetics of Roselle seeds. The conclusion is made by the result of statistical analysis done on a few selected drying models, producing evident with $R=0.999$, $\text{MBE}=2.37 \times 10^{-4}$, $\text{RMSE}=0.01335$ and $\chi^2=0.007794$. Thus, the objective is successfully achieved.
Acknowledgement
The authors would like to thank all the support from Faculty of Chemical Engineering, UiTM Cawangan Pulau Pinang including resources, necessary facilities such as laboratory and instruments, student and laboratory assistants for this research.

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