Thin Layer Drying Model of Bacterial Cellulose Film

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Abstract. The bacterial cellulose film produced by Acetobacter xylinum using coconut water as a carbon source was dried at a temperature of 60 to 100 °C. The drying process of bacterial cellulose film occur at falling rate drying period. Increasing drying temperature will shorten the drying time. The drying data fitted with thin layer drying models that widely used, Newton, Page and Henderson and Pabis models. All thin layer drying models describe the experimental data well, but Page model is better than the other models on all various temperature with coefficients of determination (R²) range from 0.9908 to 0.9979, chi square range from 0.000212 to 0.000851 and RMSE range from 0.014307 to 0.0289458.

1. Introduction

The interest in bacterial cellulose began to grow since the discovery of the unique physical properties of bacterial cellulose. Applications of bacterial cellulose use were extensive, from the fields of food, medicine, electronics and cosmetics [1–7]. Bacterial cellulose produced by the bacterial species Acetobacter xylinum has different properties than cellulose commonly found in plants. Film or sheet from bacterial cellulose has a mechanical strength that is better than film from a synthetic polymer.[8,9]. Bacterial cellulose film can be used as a skincare ingredient, facial mask, food wrapping, drug delivery, wound dressing and as the membrane material [2,6,7,9].

Previous studies have reported that the nata de coco film produced by Acetobacter xylinum contains major components of water and cellulose, so for further utilization, the bacterial cellulose needs to be dried. For practical uses, bacterial cellulose in dried form is more portable and convenient than the wet one [7].

The drying of material is a complicated process that needs much energy and time. Drying process at industrial scale consumes up to 15% total energy requirements for entire processes [10-12]. Drying of materials can increase shelf-life and reduces the bulk and mass of the material, thus simplifying storage, packaging, and transport. Sometimes, drying procedure can reduce the product volume, leading to a decrease space requirements for storage. Therefore knowledge of the drying characteristics of the material is necessary. So this research was done to study the drying nature of bacterial cellulose film and found the appropriate model to describe the characteristic drying of bacterial cellulose film.
2. Materials and methods

2.1. Materials

*Acetobacter xylinum* (strain FNC-0001) obtained from Gadjah Mada University (UGM). Coconut water was from local market at Yogyakarta, Indonesia. Yeast extract, acetic acid, sucrose, distilled water, NaOH, urea, universal pH paper, plate count agar were from Merck.

2.2. Bacterial cellulose film production

The production of bacterial cellulose adopted from Radiman and Yuliani (2008) with slight modifications [13]. The culture medium used to produce bacterial cellulose consist of 1 liter of coconut water, 50 g of sucrose, 10 g of urea, starter of *A. xylinum* (10%) and the pH is set to 5 with acetic acid. The media was then sterilized by autoclave at 121 °C for 15 minutes. Bacterial cellulose was grown at static conditions for 10 days at room temperature. After incubation, the bacterial cellulose was boiled in distilled water for 2 hours, and washed in running water until neutral.

2.3. Drying process

The bacterial cellulose film with a diameter of 3.25 cm was dried in moisture analyzer A&D MX-50 at a temperature of 60, 70, 80 and 100 °C. The drying data was collected automatically using PC with A&D WinCT-Moisture software installed.

2.4. Mathematical Modelling

The moisture ratio (MR) of the bacterial cellulose was calculated using the following equation:

\[ MR = \frac{M_e - M_t}{M_0 - M_e} \]  \hspace{1cm} (1)

where \( M_t \) is the moisture content at any time of drying, \( M_0 \) is initial moisture content and \( M_e \) is equilibrium moisture content. The experimental data fitted with widely used thin layer drying model i.e. Newton, Page and Henderson and Pabis models [12,14–19].

Newton model is also known as Exponential or Single exponential model or the Lewis model. The Newton model is the simplest thin layer model because it has only one constant.

\[ MR = \exp(-kt) \]  \hspace{1cm} (2)

where MR is moisture ratio, k constant and t is time.

The Page model is a modification of the Newton model to overcome the deficiencies of the model by adding the dimensionless empirical constants (n) at the time parameter, so this model gives better moisture loss prediction results.

\[ MR = \exp(-kt^n) \]  \hspace{1cm} (3)

where MR is moisture ratio, k, n constant and t is time.

Henderson and Pabis model is based on Fick’s second law of diffusion and widely used to predict the drying nature of biological materials.

\[ MR = a \exp(-kt) \]  \hspace{1cm} (4)

where MR is moisture ratio, a, k constant and t is time.

A widely used statistical evaluation to assess goodness of fit of the mathematical equations to the experimental data are correlation coefficient (R²), root mean square error (RMSE) and chi-square (\( \chi^2 \)) [12,16,19–22]. The correlation coefficient (R²) is widely used for selecting the best equation to describe the equation of the drying curve and the highest r value is required. Another criterion for
selecting the best model is chi-square, and root mean square error (RMSE). The chi-square value is the mean square of the deviations between the experimental and predicted values for the models and was used to determine the goodness of the fit. The lower the values of the chi-square were, the better the goodness of the fit. The RMSE value shows the deviation between the experimental and predicted values.

\[
\chi^2 = \frac{\sum_{i=1}^{N}(MR_{\text{exp},i} - MR_{\text{pre},i})^2}{N-z} \qquad (5)
\]

\[
RMSE = \left[ \frac{1}{N} \sum_{i=1}^{N}(MR_{\text{pre},i} - MR_{\text{exp},i})^2 \right]^{1/2} \quad (6)
\]

where \(MR_{\text{exp}}\) is moisture ratio from experimental data, \(MR_{\text{pre}}\) is moisture ratio prediction, \(N\) is number of data observed, \(z\) is number of constant parameter and \(t\) is time.

3. Results and Discussion

The experimental results of the cellulose drying process show that the moisture content and the drying rate decrease with the drying time. The moisture loss is high at the initial period and becomes slow in the last period of drying process. The rate of drying is also high during the initial period but becomes very slow in the last stage of the drying process. In the early period of drying, free moisture content located on the surface of the material is still high. The free water content at the surface of the material is very easily released into the environment resulting in a large decreased of moisture content from material at the beginning of drying period. At a later stage, the moisture from within the material move to the surface by diffusion resulting in a mass transfer of water from the material to the environment becomes slower [23].

![Figure 1](image_url)

**Figure 1.** Moisture ratio over time in different drying temperatures

Figure 1 shows the drying kinetic of bacterial cellulose film at different temperatures. Figure 1 indicates that the drying process becomes shorter as the drying temperature increases. Previous studies on drying of biological materials show the same result [17,20,22]. Increasing the drying temperature would increase the drying rate, so at higher drying temperature, drying process becomes faster. Increasing the drying temperature from 60 to 100 °C shortened the drying time from 664 minutes to 291 minutes.
Figure 2. Drying rate of bacterial cellulose film

Figure 2 shows the graph of the drying rate relationship over time, where the drying rate decreases over the drying time. From Fig. 2, it can be seen that the drying process of bacterial cellulose occurs in the falling rate drying period. According to previous studies, the biological materials commonly dried at falling rate drying period [11,22,24]. The drying process at a falling rate period is more noticeable at 100 °C than at lower temperature. A high drying temperature causes the process of moving moisture from the surface of the material to the environment to be faster than the moisture transfer in the material to the surface of the material. Fan et. al. (2011) found this phenomenon generally occur in material with high moisture content [17].

Table 1. Parameter of thin layer model

| Model            | 60°C  | 70°C  | 80°C  | 100°C |
|------------------|-------|-------|-------|-------|
| Newton           | k     | R²    | χ²    | RM    |
|                  | 0.005 | 0.989 | 422   | 0.037 |
|                  | 0.005 | 0.002 | 2     | 564   |
|                  | 0.989 | 0.001 | 422   | 0.037 |
|                  | 0.005 | 0.989 | 422   | 0.037 |
|                  | 0.001 | 9     | 1     | 1     |
|                  | 0.001 | 0.002 | 0.001 | 0.002 |
|                  | 0.001 | 0.990 | 9     | 1     |
|                  | 0.001 | 0.990 | 9     | 1     |
| Page             | k     | R²    | χ²    | RM    |
|                  | 0.073 | 2     | 2     | 422   |
|                  | 0.001 | 0.002 | 0.001 | 0.002 |
|                  | 0.001 | 0.986 | 9     | 1     |
|                  | 0.001 | 0.986 | 9     | 1     |
|                  | 0.002 | 0.002 | 0.002 | 0.002 |
|                  | 0.97  | 57    | 0.050 | 0.044 |
|                  | 0.073 | 57    | 0.050 | 0.044 |
|                  | 0.001 | 0.002 | 0.002 | 0.002 |
|                  | 0.001 | 0.002 | 0.002 | 0.002 |
|                  | 0.002 | 0.002 | 0.002 | 0.002 |
|                  | 0.829 | 962   | 0.044 | 0.048 |
|                  | 0.829 | 962   | 0.044 | 0.048 |
|                  | 0.001 | 0.002 | 0.002 | 0.002 |
|                  | 0.001 | 0.002 | 0.002 | 0.002 |
| Henderson &Pabis| a     | R²    | χ²    | RM    |
|                  | 48    | 85    | 85    | 927   |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.97  | 51    | 607   | 411   |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.86  | 411   | 411   | 307   |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 48    | 411   | 411   | 307   |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 411   | 307   | 307   | 307   |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 0.005 | 0.008 | 0.008 | 0.028 |
|                  | 307   | 307   | 307   | 307   |

The thin layer drying models used show good prediction for the bacterial cellulose drying process, with \( R^2 \) range from 0.9815 to 0.9979, \( \chi^2 \) range from 0.000212 to 0.002557 and RMSE range from 0.014307 to 0.050374 as shown in Table 1. The Newton, Page and Henderson and Pabis models could use to fit with experimental data very well. The Newton model has \( R^2 \) value 0.9864-0.9909, \( \chi^2 \) 0.001422-0.002557 and RMSE 0.037564-0.050374. The Page model has \( R^2 \) value 0.9908-0.9979, \( \chi^2 \) 0.000212-0.000851 and RMSE 0.014307-0.0289458. The Henderson & Pabis model has \( R^2 \) value 0.9815-0.987, \( \chi^2 \) 0.00133-0.002101 and RMSE 0.036195-0.0454795. From the fitting assessment value between experimental data and models as shown in Table 1, it's clear that Page model was better than Newton and Henderson & Pabis models. These results confirm the results of previous studies that Page equations are well suited to predict the drying of biological materials [11]. Furthermore, American Standard has adopted this model as their standard for thin layer drying model of agricultural and biological products (ANSI/ASAE 2014)[11].

4. Conclusion

The drying of bacterial cellulose film takes place in the falling drying period and is strongly influenced by temperature. The thin layer drying equation can predict the kinetic drying of bacterial cellulose very well. The thin layer drying model proposed by Page give best prediction of drying bacterial cellulose film.

Acknowledgment

This study was financially supported by Indonesian Ministry of Research, Technology, and Higher Education through INSINAS Grant 2017 No: 04/INS-2/PPK/E/E4/2017.

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