Drying kinetics of technical specified rubber

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ABSTRACT

This paper reports the study of crumb rubber drying in different experimental designs. It is important to understand the characteristics of crumb rubber drying in order to formulate a better drying strategy that could give higher energy efficiency. Four experiments were carried out with constant heat at maximum 100 °C and a stainless steel container was used to hold the sample of crumb rubber under study. The surface temperature profile of the rubber was investigated using two types of drying methods, normal hot air drying and vacuum drying. It was found that when the sample was dried, external surface temperature for drying with hot air dryer was higher than vacuum dryer. The results showed the evolution of temperature profile was not in good agreement with the prediction which revealed that there was no temperature gradient within the drying samples. The energy consumption for vacuum drying was higher compared to hot air drying, where there was a difference of 0.7079 MJ/kg H2O evaporated for drying temperature at 100 °C. The best fit model generated from the experimental data was the modified Henderson and Pabis model and the highest effective diffusivity obtained was 5.243 × 10⁻⁹ m²/s heating by vacuum oven at 90 °C under zero atmospheric pressure.

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1. Introduction

Since early twentieth century, rubber has been an important commodity for Malaysia [1]. The success in rubber planting and the fast development of automobile industries have made Malaysia one of the leading rubber exporters. Rubber latex is the sap, a protective layer beneath the bark of rubber tree (Hevea brasiliensis), which is normally tapped from the bark of Hevea Brasiliensis tree, is also named as polyisoprene. This rubber tree originated from Brazil and its seedlings were then exported to Sri Lanka, Singapore and other Asia countries, including Malaysia [2]. The early use of rubber was restricted to waterproof shoes, it was then further popularized when Charles Goodyear vulcanized the rubber into modified rubber [3].

The current main rubber products for medical industries, baby care and automotive industries are commonly produced from concentrated latex or solid rubber. Both types of rubber have to be further processed once the sap was tapped from tree. The centrifugation of field latex will be able to produce concentrated latex in liquid form with dry rubber content (DRC) of 60% and above [4]; while rubber sheet or crumb rubber is a type of dry rubber products that is rather important for tyre industries. The raw rubber that was required for tyre manufacturing process, known as crumb rubber, is commonly marketed in various grade and wrapped with polyethylene plastic sheets [5]. Crumb rubber is also known as “technical specified rubber” [6], where 70% are sold to tyre industries [7].
In this paper, the drying of technical specified rubber (TSR) was investigated. In TSR, the initial plasticity, plasticity retention index and volatile matter are very important. For related rubber products like gloves, the mechanical strength, including tensile strength, elasticity, and modulus are part of the crucial test to validate product quality. The rubber product’s properties usually remain unchanged at normal room temperature, but the strength, elasticity and flexibility of rubber deteriorate faster when stored in hot condition. Generally, crumb rubber was graded according to Standard Malaysia Rubber (SMR), where the requirement was as mentioned in Table 1.

Crumb rubber drying process is important in minimizing the moisture content in rubber, reduction of water activity, and to ensure consistent end product quality. Researches by Kulchanat [8] and Yodthong [9] have shown that the addition of wood vinegar to the drying process able to reduce bacteria growth and inhibits anti-fungal properties on rubber. Thus, the main objective of this study is to understand the drying kinetic of crumb rubber and changes of temperature profile on rubber surface when drying under hot air. The current rubber drying requires drying air temperature in the range of 130 °C and heating continuously up to three hours, but the process carries a possibility of inconsistent product quality with high energy consumption. The most popular artificial drying is the use of trolley dryer by hot air convective drying method. However, the skyrocketing fuel cost is one of the main challenges to current rubber processing plants. There are limited researches on rubber drying process. Berthomieu [10], Khongchana [11], Yutthana [12] and Suchonpanit [13] works have shown some similarities in trying to simulate and design industrial dryers. According to Khongchana [11], the specific energy consumption is measured by the equation below:

\[ SEC = \frac{2.6(\sum W_M) + (\sum Q_h)At}{W_w} \]  

(1)

where

- \( SEC \) = Specific energy consumption, MJ/kg water evaporated;
- \( Q_h \) = Heat energy consumption, kW;
- \( W_M \) = Electrical energy consumption, kW;
- \( W_w \) = Weight of water evaporated out from the rubber, kg.

However, there are multiple differences in Jutarut's [14] equation to measure the specific energy consumption:

\[ SEC = \frac{2.6(\sum E_{fan}) - (\sum Q_h)At}{M_w} \]  

(2)

Where

\( E_{fan} \) = Rate of Electrical energy consumption of fan, kW;
\( M_w \) = Weight of water evaporated out from the rubber, kg.

The calculation presented by Jutarut involved the rate of electrical energy and heat energy consumption. From the two equations mentioned, we can see obvious differences in both, whereby one was calculated based on the addition of heat energy and electrical energy, and other SEC was using subtraction of heat energy for calculation. The differences in calculation methods may lead to difficulties to compare the SEC values from different researchers. Therefore, there is a need to investigate the actual energy consumption in similar drying system. Besides evaluation of drying kinetic and specific energy consumption (SEC) of rubber drying process, a total of eleven types of mathematical models were analyzed to determine whether the drying technique was suitable for industrial scale rubber processing.

### 2. Materials and methods

#### 2.1. Raw materials and equipment

Fresh crumb rubber (Lien Rubber (M) Sdn. Bhd., Port Klang, Malaysia) was acquired from a local natural rubber processing company. The crumb rubber was coagulated and washed prior to purchase; however, further removal of dirt was necessary for higher accuracy of test. The rubber size was reduced by creper shredder machine and the appearance was similar to a long thread with diameter of 4 mm ± 1 mm. The rubber would entangle and stick to each other upon heat treatment. Therefore, proper selection of rubber, washed off surface adhering dirt, and size are important for the accuracy of drying test.

The drying of rubber was carried out by two types of dryers, universal lab oven (Memmert, UFB 500, Germany, Evergreen Engineering & Resources) and vacuum oven (Tuff, TVAC-92, Germany, Tech-Lab Scientific Sdn Bhd) with the use of a stainless steel container 150 mm × 75 mm × 75 mm made by Sphere Corporation. A schematic diagram (see Fig. 1) showed how the rubber sample was placed into the container. The universal oven employed hot air (HA) drying method by natural convection, with a heater of 1600 watt and temperature accuracy of ±0.1 °C; while the vacuum dryer employed vacuum drying (VD), which consisted of a build in

<table>
<thead>
<tr>
<th>Parameter</th>
<th>SMR 10</th>
<th>SMR 20</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dirt Retained on 44 Aperture (Max, % wt)</td>
<td>0.08</td>
<td>0.16</td>
</tr>
<tr>
<td>Ash Content (Max, % wt)</td>
<td>0.75</td>
<td>1.00</td>
</tr>
<tr>
<td>Volatile Matter (Max, % wt)</td>
<td>0.80</td>
<td>0.80</td>
</tr>
<tr>
<td>Nitrogen Content (Max, % wt)</td>
<td>0.60</td>
<td>0.60</td>
</tr>
<tr>
<td>Wallace rapid plasticity, P0 (Min, %)</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>Plasticity retention index, PRI (Min, %)</td>
<td>50</td>
<td>40</td>
</tr>
</tbody>
</table>
vacuum pump, temperature controlling unit, and a heater of 2400 watt with accuracy ±0.1%. An analytical weighing balance (Mettler Toledo, ME204, Malaysia, Mettler-Toledo (M) Sdn Bhd) with accuracy up to ±0.0001 g was utilized to measure the weight changes of rubber compound throughout the experiment, while the drying time for all experiments were determined by using a digital sport timer (Avantec, TM-104, Malaysia, Suria Pembekal Umum Sdn Bhd).

2.2. Process control and design

The crumb rubber sample was thoroughly cleaned and soaked in cold water for ten minutes prior to drying to obtain a saturated rubber sample. Then, the rubber was cut into small pieces to allow uniform heat distribution on the samples, and to fit into the stainless steel container. The rubber sample subjected to drying had a dimension of (average length, width and height) 150 mm × 75 mm × 30 mm. The mass of the rubber was determined before drying and throughout the heating process with increasing interval period. The rubber samples were heated at a pre-set drying temperature in the range of 80–100°C and the process was continuously monitored up to 65 min. Uniformity in drying was maintained throughout the study to determine the effect of heating temperature on rubber, including weight change and moisture content. The dried samples were then cooled to room temperature after each drying experiment, and sealed in polyethylene bags for future references. The influence of dryer on temperature profile and specific energy consumption was determined from the experiment carried out. Some measurements were taken prior to drying such as initial moisture content and mass of each sample at 65% (dry basis) and ~30 ± 5 g, respectively. The moisture content of fresh sample and dried sample were obtained by the AOAC method No. 934.06. The average ambient room temperature and relative humidity were recorded at 27°C and 80%, respectively.

2.3. Moisture content, moisture ratio and drying rate

In order to understand the fundamental drying process in rubber, the measurement of pre-set drying temperature, ambient room temperature, rubber’s surface temperature, and initial moisture content were made. The average initial moisture content in rubber was 65 ± 1%, while wet basis moisture was 39 ± 1% and the data was continuously recorded for 65 min. There were four drying conditions being evaluated as shown in Table 2. The dry basis moisture content, MC was calculated by using the below equation:

\[
\text{Moisture Content (grams water/gram dry solid)} = \frac{\text{Weight of water}}{\text{Weight of dry solids present}} \quad (3)
\]

During the drying process, all temperature data were measured with an infrared thermometer, with an accuracy of ±2°C. The drying rate was determined from the changes of mass with time, while drying constant, \( k \) was typically obtained from the slope of the negative natural log of the moisture ratio, ln (MR) versus time. The moisture ratio, MR was calculated from the results obtained by following Fick’s second law [16] in the theoretical model of thin layer drying:

\[
\text{MR} = \frac{M - M_e}{M_i - M_e} \quad (4)
\]

Where MR is moisture ratio, \( M_i \) is initial moisture content, \( M_e \) is equilibrium moisture content, and \( M \) is moisture content at time \( t \).

The drying kinetic and drying rate for rubber drying were calculated based on the data obtained. The drying rate, \( W \) (g H₂O/m² min) was defined by:

\[
W = -\frac{W_d}{A} \left(\frac{dX}{dt}\right) \quad (5)
\]

where \( W \) is drying rate, \( W_d \) is weight of dry solid (Bone dry mass), \( A \) is the contact surface of the drying gas and the rubber, and \( dX/dt \) is the humidity variation (dry basis) over time \( t \).
2.4. Specific energy consumption (SEC)

SEC denoted the energy required to remove a unit mass of water. For the calculation of SEC, the equation used as reference is the equation from the study of Bualuang [17]. The calculation based on this equation could adequately compare the SEC of two different dryers, while the two equations of Khongchana [11] and Jutarut [14] mentioned earlier can only calculate the SEC based on water evaporated, heat energy and electrical energy consumption without considering the bone dry mass of sample.

\[
SEC = \frac{3.6P}{(M_i - M_f)W_d}
\]

where \(P\) is power, \(M_i\) is initial moisture content, \(M_f\) is final moisture content; and \(W_d\) is bone dry mass of sample, kg.

2.5. Drying kinetic models

By evaluating the drying kinetic of rubber, the efficiency of vacuum drying and hot air convective drying method was analyzed. The analysis allowed the determination of suitable drying technique that was suitable for industrial scale rubber processing. There were eleven types of mathematical models to describe the drying kinetics of rubber (see Table 3). Some of the earlier models were based on thin layer rapid drying with low temperature heating profile, which was not suitable for applying on thick layer drying experimental works [18]. The complex partial differential equation that involved mass balance, drying rate, heat balance and transfer would be more accurate, but a model must be validated by comparison with experimental results. The model that was able to describe rubber drying characteristics would give the best fit on the experimental data. The correlation coefficient (R²), chi-square and RMSE were used to determine the quality of the fit. The highest value of R², where the value greater than 0.995 indicating a good fit. The model with lowest values of chi-square and RMSE were chosen as the best empirical model equation for rubber drying.

3. Results and discussions

3.1. Drying curve

From the rubber drying studies, the temperature profile through changes of rubber’s surface temperature (see Fig. 2) was investigated. For initial heating process, the heat transfer was fast as the moisture content of rubber was high. As the evaporation process of water was exothermic, heat was absorbed quickly by the rubber when both tests were initiated under setting temperature 100 °C. For drying under vacuum condition, the slow rising of rubber’s surface temperature indicated that the heat conduction to the rubber sample was not effective. As rubber was not a good heat conductor, the conduction of heat through contact surface would be slow. The highest rubber surface temperature measured throughout the test was only 74 °C, which indicated that there was heat loss during thermal diffusion in the rubber. However, a total of 10.998 g moisture was able to be removed due to the intense heat provided by the vacuum oven compared to universal oven. For the drying test carried out by universal oven, the dryer provided sufficient air flow to spread the heat evenly on rubber surface, with higher impact on rubber’s surface temperature compared to vacuum drying, but the total moisture loss was 8.091 g only. The drying in vacuum condition was faster compared to hot air drying by

<table>
<thead>
<tr>
<th>Code</th>
<th>Heat source</th>
<th>Drying temp. (°C)</th>
<th>Drying time (min)</th>
<th>Atmospheric pressure (atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>T1</td>
<td>Vacuum oven</td>
<td>80</td>
<td>65</td>
<td>0</td>
</tr>
<tr>
<td>T2</td>
<td>Vacuum oven</td>
<td>90</td>
<td>65</td>
<td>0</td>
</tr>
<tr>
<td>T3</td>
<td>Vacuum oven</td>
<td>100</td>
<td>65</td>
<td>1</td>
</tr>
<tr>
<td>T4</td>
<td>Universal oven</td>
<td>100</td>
<td>65</td>
<td>1</td>
</tr>
</tbody>
</table>

*T4 was conducted to imitate current industrial drying by hot air convective method [15].

<table>
<thead>
<tr>
<th>Model No.</th>
<th>Model name</th>
<th>Model equation</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Lewis</td>
<td>MR = exp(−kt)</td>
<td>Bruce (1985)</td>
</tr>
<tr>
<td>2</td>
<td>Page</td>
<td>MR = exp(−kt&lt;sup&gt;a&lt;/sup&gt;)</td>
<td>Page (1949)</td>
</tr>
<tr>
<td>3</td>
<td>Logistic</td>
<td>MR = a/(1 + exp(−kt))</td>
<td>Apintananapong (2009)</td>
</tr>
<tr>
<td>4</td>
<td>Two term</td>
<td>MR = a exp(−kt) + b exp(−k&lt;sup&gt;2&lt;/sup&gt;t)</td>
<td>Henderson (1974)</td>
</tr>
<tr>
<td>5</td>
<td>Wang and Singh</td>
<td>MR = 1 + at + bt&lt;sup&gt;2&lt;/sup&gt;</td>
<td>Wang and Singh (1978)</td>
</tr>
<tr>
<td>6</td>
<td>Henderson and Pabis</td>
<td>MR = a exp(−kt)</td>
<td>Henderson and Pabis (1961)</td>
</tr>
<tr>
<td>7</td>
<td>Logarithmic</td>
<td>MR = a exp(−kt) + c</td>
<td>Yaldız et al. (2001)</td>
</tr>
<tr>
<td>8</td>
<td>Approximation of diffusion</td>
<td>MR = a exp(−kt) + (1 − a) exp(−kbt)</td>
<td>Yaldız et al. (2001)</td>
</tr>
<tr>
<td>9</td>
<td>Two term exponential</td>
<td>MR = a exp(−kt) + (1 − a) exp(−kat)</td>
<td>Akpinar et al. (2003)</td>
</tr>
<tr>
<td>10</td>
<td>Modified Henderson and Pabis</td>
<td>MR = a exp(−k&lt;sup&gt;2&lt;/sup&gt;t) + b exp(−k&lt;sup&gt;3&lt;/sup&gt;−c)</td>
<td>Tasara et al. (2011)</td>
</tr>
<tr>
<td>11</td>
<td>Hii model</td>
<td>MR = a exp(−kt&lt;sup&gt;n&lt;/sup&gt;) + c exp(−kt&lt;sup&gt;3&lt;/sup&gt;)</td>
<td>Hii et al. (2009)</td>
</tr>
</tbody>
</table>

Note: k, a, b, c, g and n are drying constants.
referring to the moisture removal rate and calculated drying rate (see Table 4).

For moisture removal according to time interval, the water removal rate was rapid in the first ten minutes and decelerated as moisture content started to reduce. As the surface moisture dried out, the moisture need longer time to diffuse out from rubber. The stickiness of the rubber inhibited the diffusion of internal moisture to rubber surface, but the heat would constantly soften the rubber, making it more compact as compared to the time when it was moist. The changing of moisture ratio (MR) versus time (see Fig. 3) showed that MR decreased continuously throughout 65 min and no constant drying rate period existed. It is apparent that the drying became slower after the first drying period and falling-rate period was present. As the moisture was removed from rubber, the rubber got thinner and experienced slight shrinkage throughout the drying period, it was predicted that the drying condition (including humidity, temperature and air flow) throughout the rubber were constant.

Drying kinetics was studied for moisture contents 65–20% (w/w). Four drying curves of rubber with drying temperature of 80–100°C versus moisture content were plotted (see Fig. 4). All drying condition showed a trend of reducing drying rate, this was due to the increase of rubber’s bulk density and the reduction of pore for air flow. The initial moisture content (MC) of all samples were maintained at 65% and all tests were able to get results lower than 20% dry basis MC within 65 min. This result showed that the rubber would be able to dry to desired moisture content at shorter given time, instead of three hours as per industrial practice. From Fig. 4, it can be seen that falling-rate period began after the warm up time and subsequent drying process by universal oven was slower as compared to vacuum oven. The experimental work done by using universal oven also obtained results with higher MCf compared to vacuum oven.

### 3.2. Specific energy consumption (SEC)

The results demonstrated that the drying rate by vacuum oven at 100°C was highest among all the four tests (see Table 4) even though the drying process was operated at 1 atmospheric pressure. This showed that rubber was more suitable to dry in a condition with hot air as medium compared to in vacuum state. However, the SEC results showed that universal oven had lowest MJ/kg H2O evaporated, the energy consumption for vacuum dryer was higher compared to hot air dryer, which agreed to same findings on other research work for different materials [19].

<table>
<thead>
<tr>
<th>Exp</th>
<th>Heat Source</th>
<th>Drying temp. (°C)</th>
<th>Drying time (min)</th>
<th>Average drying rate (g H2O/m2 min)</th>
<th>Deff (m2/s)</th>
<th>P (kW-h)</th>
<th>SEC (MJ/kg H2O) evaporated</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Vacuum oven</td>
<td>80</td>
<td>65</td>
<td>15.8437</td>
<td>3.716 × 10⁻⁹</td>
<td>9.360</td>
<td>9.1095</td>
</tr>
<tr>
<td>2</td>
<td>Vacuum oven</td>
<td>90</td>
<td>65</td>
<td>13.4385</td>
<td>5.243 × 10⁻⁹</td>
<td>9.360</td>
<td>9.7095</td>
</tr>
<tr>
<td>3</td>
<td>Vacuum oven</td>
<td>100</td>
<td>65</td>
<td>16.5931</td>
<td>4.537 × 10⁻⁹</td>
<td>9.360</td>
<td>8.5103</td>
</tr>
<tr>
<td>4</td>
<td>Universal oven</td>
<td>100</td>
<td>65</td>
<td>10.3529</td>
<td>3.830 × 10⁻⁹</td>
<td>6.239</td>
<td>7.8024</td>
</tr>
</tbody>
</table>
3.3 Drying kinetic models

From the comparison of eleven mathematical models mentioned, modified Henderson and Pabis Model was the most suitable empirical model (see Table 5), as it had high $R^2$ value of minimum 0.9979 and lowest chi-square value of maximum 0.002. The arbitrary constant for the drying condition was as described (see Table 6). This empirical modified Henderson and Pabis model has shown a better fit to the experimental rubber drying data as compared to others models. Consequently, the modified Henderson and Pabis drying model could adequately describe the drying behavior of rubber, a good correlation between the values of model and experiment data was found. The predicted moisture ratio versus experimental moisture ratio graph for four drying condition was presented in four charts (see Fig. 5). From the slope of graph ln (MR) versus time, the value of effective moisture diffusivity was calculated and the value of $D_{eff}$ for all four drying condition were presented in Table 4.

4. Conclusions

The drying curve revealed that rubber drying process involved multiple stages. After the initial warm up period, the drying process continued in falling rate period and no constant rate
period existed. The falling rate was due to the shrinkage of material and increase in bulk density. The study also revealed that the drying rate changes with different dryers’ performances, whereby hot air drying method consumed less heat energy with less moisture removal in the process. The specific energy consumption of hot air drying showed that it can reduce the energy consumption for rubber drying. However, more heating time was required when hot air drying method was employed in the drying process.

The temperature profile of rubber showed the effectiveness of heat transfer from surrounding to rubber. The universal oven was able to distribute heat faster compared to vacuum oven. The drying kinetics was mainly affected by drying temperature, where higher temperature improved the drying rate and reduction of moisture content in rubber sample. The drying rate was higher in vacuum condition due to the condition of lower atmospheric pressure, so drying in vacuum condition was able to reduce MC in higher rate. The empirical model named modified Henderson and Pabis model were the best fit model for the measured experimental values.

![Graphs showing moisture ratio comparison](image)

**Table 6 – Arbitrary constant of four drying condition by modified Henderson and Pabis Model.**

<table>
<thead>
<tr>
<th>Arbitrary constant/drying condition</th>
<th>Henderson and Pabis Model</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>T1–VA 80 °C</td>
</tr>
<tr>
<td>k1</td>
<td>0.005690974</td>
</tr>
<tr>
<td>a</td>
<td>0.376967754</td>
</tr>
<tr>
<td>b</td>
<td>0.503064087</td>
</tr>
<tr>
<td>k2</td>
<td>0.491953271</td>
</tr>
<tr>
<td>c</td>
<td>0.127525422</td>
</tr>
<tr>
<td>k3</td>
<td>0.043057648</td>
</tr>
</tbody>
</table>

**Fig. 5 – Comparison of moisture ratio between experimental data and calculated values according to modified Henderson and Pabis model (a) vacuum oven 80 °C; (b) vacuum oven 90 °C; (c) vacuum oven 100 °C; (d) universal oven 100 °C.**

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