

Mathematical Models of Natural Rubber Sheets Drying: Difference Acid Coagulation Cases

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ARTICLE INFO	ABSTRACT
Article history: Received 28 November 2023 Received in revised form 24 April 2024 Accepted 5 May 2024 Available online 30 May 2024 <i>Keywords:</i> Natural rubber sheets; moisture	The mathematical model for drying process is a useful tool in process optimization and drying chamber design. The research purposes of this study were to investigate the influence of drying temperature on drying time and the modelling the drying kinetics of the natural rubber (NR) sheets. The NR sheets which produce from commercial formic acid, commercial acetic acids, and ammonia plus commercial formic acid were studied at drying temperature of 40, 50, and 60°C and air speed of 0.5 m/s. The results indicated that the drying time was substantially reduced with an increase in temperature. The moisture content ratio of rubber sheets produce from commercial formic acid coagulation was similar to the sheets produce from commercial acetic acids coagulation. However, the drying time of them were longer than the drying time of the sheets produce from ammonia plus commercial formic acid coagulation. Finally, the logarithmic model was the best model which suitable to predict the moisture content ratio of the sheets drying with
content; mathematical models	all experimental condition.

1. Introduction

Thailand is well-known as among the biggest natural rubber (NR) production and exportation country in the world. In 2020, Thailand Rubber Research Institute (TRRI) has recorded the total amount of NR production of Thailand is about 4.2 million metric tons [1]. The mono-graph of TRRI shows NR in Thailand is produced in four types, namely, ribbed smoked sheets (RSS), block rubber (various grades of Standard Thai Rubber, STR grades), rubber concentrated latex, and other forms (rubber blend composite and other). NR is the raw material used in the production of gloves, medical devices, automotive tire, couplings and conveyor belts, etc. [2-5]. In the world's rubber market, the largest consumers are USA, China, Japan and India, while China and Japan are Thailand's largest exporting markets. As is known, the ribbed smoked sheets (RSS) and air-dried sheet (ADS) rubber is one of the major forms of NR products [6]. The ADS rubber process has the same procedure as RSS

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production except for the fuel used to dry the NR sheets. In RSS process, hot gas is carried from the combustion of firewood species such as para rubber, eucalyptus, acacia, etc. [5,7]. While the ADS rubber process, hot gas is carried from the heater. Currently, the worldwide consumption of RSS and ADS rubber increased due to increasing demands of the tire industry. In the ADS rubber process, rubber sheets are washed in a pool and hung on the bamboo bars to dry with ambient air about 6-12 hours before transferring to the drying chamber. Hot gas from heaters flowed to the rubber sheets for dehydration their moisture content by the blower [5,8]. The ADS product of Thailand comes from the small-level community-based on the cooperatives [5,9,10]. So, the quality of them is not constants. This problem has negative effects for their prices. The drying process of ADS consumes most of the energy in moistures obliteration process and the drying quality will surely be affected by the efficiency of the chamber [11]. The moisture must be reduced to the grade passable for storage and marketing of ADS. Thus, it is essential to control the drying conditions precisely in order to improve the product quality and reduce energy consumption in the drying process [11-13]. In the chamber, the sheet samples are hung on bamboo bars until they are evenly dried. In order to reduce this problem, study and improvement of the mathematical models for drying process should been required.

Many researchers studied the mathematical model for the drying kinetics of food products [14-21]. However, there are very few reports on the mathematical model changes of NR sheets drying and its composites [22,23]. As is known, the NR sheet color change in the non-enzymatic and enzymatic browning reactions are related to the drying time and temperature. The color value increased with increasing the drying temperature and drying time. Kalasee and Dangwilailux [24] have been the only ones studying the effects of the chemical and physical properties when coagulating with different acetic acid. Their paper predicts the color changes of the drying and color kinetics of NR sheets during the drying process. Thus, the purpose of this research was to study the mathematical model changes for the drying kinetics of natural rubber sheets which were related to the coagulating acid type, commercial formic acid, commercial acetic acids, and ammonia plus commercial formic acid during drying process. The models on the experimental data of moisture ratio values using non-linear regression method were estimated at temperatures 40, 50, and 60°C and air speed of 0.5 m/s.

2. Methodology

2.1 Materials

For preparing rubber sheets, the standard of Rubber Authority of Thailand (RAOT), fresh natural rubber latex to be tested is tapped and collected from RRIM 600, type of Para-rubber tree. It was obtained from the Agriculture, Food and Energy Center of King Mongkut's Institute of Technology Ladkrabang from the Prince of Chumphon Campus, Pathio District, Chumphon Province, Thailand. Latex was diluted with water and coagulated by addition of the acids; commercial formic acid, commercial acetic acids, and ammonia plus commercial formic acid; in the coagulating tank. After storage about two hours, the slabs were compressed between two rollers and then washed with clean water and hung on bamboo bars before drying at the chamber. In drying process, NR sheets were carried on the chamber which set-up inlet air velocity and the temperature until the final moisture content decreased to $3.0\pm1.0\%$ dry basis [8,13,24]. In this study, the $0.6 \times 0.8 \times 1.0$ m in size (width × length × height) hot air oven chamber with containing 100 kg of NR sheets was investigated, as shown in Figure 1. This chamber was constructed of steel. A 1.5-kW heater (Maxthermo, model MC-2438, Taiwan) and a centrifugal blower (Nitco, model RB60-520, Hessdorf, Germany) driven by a

2.2-kW motor were also used as the temperature and airspeed controllers, respectively. Moreover, the sample size of NR sheet was 0.3×0.1 m and thickness 3 mm.



natural rubber sheets

2.2 Experimental Procedure

In the experimental, the NR sheets were carried on the chamber which set-up the drying temperature of 40, 50, and 60°C and air speed of 0.5 m/s. These values are estimated and considered from simulation data with an air velocity inlet into the NR sheets drying chamber by computational fluid dynamics (CFD) modified from Dejchanchaiwong *et al.*, [3], and Promtong and Tekasakul [25]. The initial moisture content of NR sheets was 80.0±2.0% dry basis and the final moisture content was less than 3.0% dry basis, respectively. The ambient air temperature, inlet and outlet drying air temperature, and the grain temperature was measured by K-type thermocouples. The moisture contents of the sample natural rubber sheets were calculated by standard method of the Association of Official Analytical Chemists (AOAC) [26].

2.3 Data Analysis

The drying models improved from experimental data at different drying temperatures were shown in Table 1. The best empirical model of NR sheets was chosen as the one with the lowest mean relative deviation (MRD) value and the highest coefficient of determination (r^2) value. For each experiment point, MRD was determined by Eq. (1).

$$MRD = \frac{1}{n} \left(\sum_{i=1}^{n} \frac{\left| M_{R,i} - \dot{M}_{R,i} \right|}{M_{R,i}} \right)$$
(1)

where $M_{R,i}$ is the measured valve data, $\dot{M}_{R,i}$ is the estimated valve through fitting of the drying model, and n is the number of experimental points.

According to the drying models, the dimensionless moisture ratio (M_R) was given by Eq. (2).

$$M_{R} = \frac{M - M_{eq}}{M_{int} - M_{eq}}$$
(2)

where M, M_{eq} , M_{int} are the moisture content at any time, the equilibrium moisture content, and the initial moisture content in dry basis percentage, respectively.

The mathematical models for drying process							
Model	Equation*	Reference					
Page	$M_p = a \cdot \exp(-k \cdot t)$	Babalis <i>et al.,</i> [15],					
		Mazutti <i>et al.,</i> [17],					
		Janjai <i>et al.,</i> [18]					
Logarithmic	$M_{\rm p} = a \cdot \exp(-k \cdot t) + b$	Akpinar <i>et al.,</i> [14],					
	R = R	Babalis <i>et al.,</i> [15],					
		Mazutti <i>et al.,</i> [17],					
		Janjai <i>et al.,</i> [18]					
Two-term exponential	$M_{\rm p} = a \cdot \exp(-k \cdot t) + (1 - a) \exp(-k \cdot a \cdot t)$	Wang <i>et al.,</i> [16],					
	$= \frac{1}{K} = $	Janjai <i>et al.,</i> [18]					

 *M_R is the moisture ratio (dimensionless) and a, b, and k are the equation constants of the mathematical models for drying kinetics of the NR sheets produce

3. Results

Table 1

3.1 Influence of Drying Temperature on Drying Time

For all the experiments, the NR sheets having an initial moisture content of dry basis were dried to reach a final moisture content of dry basis. The influence of the drying temperature on the sheet samples at each of three coagulating acid types, namely commercial formic acid, commercial acetic acids, and ammonia plus commercial formic acid, was shown in Figure 2, Figure 3 and Figure 4, representing the drying time versus the moisture ratio. It is clearly evident that the drying time increased, the moisture ratio decreased continuously. At NR sheet samples which used coagulating by commercial formic acid in the ADS rubber production, the increase in temperature decreased the drying time which needed to reach moisture content of about dry basis as same as commercial acetic acids, and ammonia plus commercial formic acid, respectively. This result was due to the increased energy of water and the space between the rubber molecules being decreased after water molecule evaporation when the temperature was increased. This agreed with the results reported in Rattanamechaiskul et al., [4], Kalasee and Dangwilailux [24], and Tarigan et al., [27]. For the effect of the type of coagulating material on the drying time; when the drying process started, the samples moisture which used coagulating by commercial formic acid and acetic acids rapidly decreased through convection heat transfer over the first 12 hour. This result was different from the sheets which used coagulating by ammonia plus commercial formic acid, when these samples moisture very rapidly decreased over the first 8 hour. Afterward, the reduce in moisture of the NR sheets was at a constant rate due to heat diffusion until drying finished at a final moisture of dry basis [24,25]. For this reason, as the drying temperature was increased, the drying time reduced.



Fig. 2. Comparison of experimental data and predicted moisture content ratios of the natural rubber sheets produce from commercial formic acid coagulation by fitting of Logarithmic model



Fig. 3. Comparison of experimental data and predicted moisture content ratios of the natural rubber sheets produce from commercial acetic acid coagulation by fitting of Logarithmic model



Fig. 4. Comparison of experimental data and predicted moisture content ratios of the natural rubber sheets produce from ammonia plus commercial formic acid coagulation by fitting of Logarithmic model

3.2 Modelling the Drying Kinetics of the Samples

The average moisture ratio of NR sheets dried was test verified with three different drying models to find out their suitability to describe the drying process. Table 2 presents the mathematical model parameters related to the NR sheets drying kinetics which used coagulating by commercial formic acid in the ADS rubber production. The results showed that the logarithmic model was suitable to predict the moisture content ratio of the natural rubber sheets drying, confirming by the mean of the coefficient of determination (r^2) and the mean of relative deviation (MRD). It showed the r^2 value was greater than 0.99 and the MRD value was less than 0.03, which is satisfactory. This result was similar to the results of the sheets produce from commercial acetic acids coagulation and ammonia plus commercial formic acid coagulation. Therefore, the logarithmic model was suitable to predict the moisture content ratio of the NR sheets drying for all samples. Although, the sheet samples which used coagulating by ammonia plus commercial formic acid had the shortest drying time about 32 hours for the drying temperature of 60°C and air speed of 0.5 m/s, an increase in drying temperature and drying time had the influence on chemical and physical properties of the NR sheets [24]. Furthermore, not only the type of coagulating material and drying temperature but also other variables that should be considered, such as the NR latex concentration, the acid concentration values, airspeed, atmosphere relative humidity, pressure, and others.

Parameters of the mathematical models for drying kinetics of the natural rubber sheets produce												
Model	Parameters	Commercial formic			Commercial acetic			Ammonia plus commercial				
	and good-	acid			acid			formic acid				
	ness of fit	Drying temperature			Drying temperature			Drying temperature				
		40°C	50°C	60°C	40°C	50°C	60°C	40°C	50°C	60°C		
Page	а	1.165	1.033	1.146	1.124	1.084	1.027	1.083	1.058	1.031		
	k	0.026	0.050	0.048	0.033	0.047	0.078	0.045	0.058	0.089		
	r ²	0.990	0.992	0.990	0.991	0.994	0.985	0.983	0.986	0.987		
	MRD	0.021	0.027	0.033	0.019	0.025	0.042	0.042	0.039	0.044		
Logarithmic	а	1.085	1.041	1.130	1.068	1.059	1.093	1.121	1.082	1.067		
	b	-0.064	-0.072	-0.146	-0.068	-0.077	-0.253	-0.234	-0.117	-0.097		
	k	0.040	0.045	0.054	0.042	0.049	0.043	0.033	0.052	0.076		
	r ²	0.998	0.997	0.998	0.999	0.998	0.995	0.995	0.994	0.991		
	MRD	0.016	0.018	0.016	0.012	0.016	0.026	0.025	0.027	0.026		
Two-term	а	0.013	0.142	0.023	0.047	0.026	0.026	0.066	1.491	0.026		
exponential	k	4.005	3.858	3.239	14.291	12.961	2.284	11.776	0.081	3.569		
	r ²	0.981	0.992	0.986	0.991	0.988	0.985	0.978	0.987	0.982		
	MRD	0.030	0.028	0.041	0.026	0.037	0.042	0.048	0.038	0.048		

Table 2



 r^2 is correlation coefficient and MRD is mean relative deviation.

4. Conclusions

The results of this research presented that an increase in drying temperature had the influence of decreasing the drying time. The moisture content ratio of the NR sheets rapidly decreased when the drying temperature was increasing. The results of the three different types of drying models showed that logarithmic model was suitable to predict the moisture content ratio of the NR sheets drying with all experimental condition. The moisture content ratio of the sheets produce from commercial formic acid coagulation was similar to the sheets produce from commercial acetic acid coagulation. However, the drying time of them are longer than the drying time of the sheets produce from ammonia plus commercial formic acid coagulation. Recommendations, in the future work, the results of this paper will be used to analyse factors and components of the rubber sheets for development the product quality and reduce energy consumption in the drying process.

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