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Artificial Neural Network Approach on Equilibrium Moisture Content for Predicting Kinetics of Air Dried Sheet Rubber

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Abstract – The objectives of this research were to predict equilibrium moisture content of fresh rubber using an artificial neural network model (ANN) and investigate Air Dried Sheet (ADS) rubber using hot air (HA) drying, green house (GH) drying and conventional open-air (OA) drying to evaluate the appropriate thin-layer drying equation model for predicting the drying kinetic of rubber sheet. Equilibrium moisture content (EMC) of natural rubber (NR) were evaluated using the gravimetric-static method among surrounding temperature of 40-60°C provided to relative humidity surrounding of 10-90%. The experimental results of equilibrium moisture content were mathematical simulated by ANN. Temperature, relative humidity were considered as the input variables to the topology of neural network. The best results of the Feed Forward Back-Propagation algorithm (FFBP) with LM algorithm (trainlm learning function) was tansig-tansig-purelin transfer function and 2-4-2-1 topology, which found to be the most suitable for describing the relationship among equilibrium moisture content, relative humidity and temperature. The appropriate thin-layer drying equation model for predicting the drying kinetic of rubber sheet. The experiments were carried out under the conditions of constant drying temperature 30-55°C, the initial moisture contents of sample rubber were operated in range of 15-40 % dry-basis. The final moisture content for each drying strategies was fixed at 1±0.05 % dry-basis. The effects of drying conditions on evolution of moisture content, drying rate and quality of rubber were determined. The experimental results showed that for both forced and natural convection the drying rate of rubber sheet was relatively related to drying temperature. The fresh rubber sheets were dried by natural sun drying took longer drying period than those of HA drying. Twelve thin layer drying equation models were used for evaluating experimental data and the results showed that the simulated data using Weibull Distribution model had a good relation to the experimental values of sample drying with HA, experiment results using Modified Henderson and Pabis model was the best fitting model for GH and conventional OA drying. According to the quality and energy analysis, the results showed that quality of the rubber sheet was acceptable in market level for all drying heat sources. However, ADS rubber drying with HA and OA were slightly better than GH drying.

Keywords: *Drying kinetic model, Hot air convection, Rubber sheet drying, Solar drying, Natural rubber*

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1. Introduction

Thailand is the largest natural rubber (NR) producing and exporting country in the world. Total amount of NR production in year 2009 about 3 million tons was continuously increased 7.07% from year 2004. The NR produced was exported about 86.15%. Generally, the natural rubber has been produced in four types; Ribbed smoked sheet (RSS), block rubber, rubber concentrated latex, and other. The mostly proportion of natural rubber produced is 31% in RSS. [1]. Like most agricultural products including all natural rubber export is hygroscopic and the storage environment could adversely affect its quality.

Tasara J. et al. [4] revealed that the equilibrium moisture content of NR using the gravimetric-static method at temperature of 40, 50, 55, and 60 °C, all of the salt solution provided relative humidity surrounding of 10-90%. The six models are Oswin, Henderson. Smith, Halsey and GAB model which using to fit the experimental data. A nonlinear regression-analysis method was used to evaluate the constants of equation. The results indicated that The Modified Oswin model was found to be the most suitable foe describing the relationship among equilibrium moisture content, relative humidity and temperature. The moisture content decreased with increasing temperature. The drying rate increased with increase of inlet air temperature and the experimental results was predicted by the Modified Henderson and Pabis thin layer drying model which was the best fitting model for describing infrared drying behavior of rubber.

Teeboonma U. et al. [14] reported the drying of ginger using infrared-vacuum technique. The cylindrical dryer, diameter 32 cm. height 30 cm. were operated on the following conditions: at drying temperature of 40, 50 and 60 °C, and absolute pressure in drying chamber of 5, 10 and 15 kPa for infrared intensity of 500 W. The ginger was dried from initial moisture content of 990-1020% dry-basis to final moisture content 10% dry-basis. It was found that the increasing of drying time or decreasing of absolute pressure in drying chamber which increasing drying rate and low specific energy consumption. The thin layer equation used to described infrared drying behavior of ginger. Experimental results using the Modified Henderson and Pabis was the best fitting model.

The objectives of the research are to application of the ANN method for describing the equilibrium moisture content behavior. and therefore to obtain the experimental data on moisture ratio and drying time of rubber sheet at different methods to dried, so as to using statistical analyzes on the mathematics models for predicting the drying kinetic of rubber sheet drying. Develop HA and GH drying system for ADS drying compared with OA.

2. Materials and Methods

2.1 *Material*

The fresh natural rubber sheets were provided from Cooperative rubber station (CRS) at Hat Yai, Songkhla province and Biochemistry research laboratory (BRL), Faculty of Science Prince of Songkhla University. The dimension of sample rubber sheet of CRS and BRL was about of 40×90 cm2 and 70×50 cm2, respectively correlated to their weight of 0.8-1.5 kg and 1.4-2.0 kg. The moisture content of samples was determined following by ASAE, 1982 method [2].

2.2 *Equilibrium moisture content (EMC)*

Equilibrium moisture content of natural rubber were determined experimentally using the gravimetric-static method The six selected salt solutions for taking equilibrium stage as followed: KNO_3, NaCl, $Mg(NO_3)_2{\bullet}6H_2O$, $MgCl_2{\bullet}6H_2O$, and LiCl. All of the saturated salt solutions provide relative humidity surroundind of 10-90% among surrounding temperature of 40-60°C. Thus, small pieces of rubber sheet samples were kept in airtight vials were tempered in incubator with a constant temperature of 40, 50, 55 and 60 °C. After a few weeks, the samples were weighted at 5 days interval until three successive reading were each less than 0.5% of the previous one. Then the moisture content of rubber sheet samples was determined by following of ASAE 1982 method [1]. The average EMC value of sample was showed by means of triplication. Finally, the experimental data were fitting curved by Artificial Neural Network (ANN) [9].

2.3 *Structure of neural network model*

Artificial neural network (ANN) model are heuristic model, same to the vast network of neurons in the human brain. The ANN model consists of several layer of interconnected neurons, which have been related with special arrangement. In this research, The network of Multilayer Feed Forward Neural Network were utilized.Also the Back-Propagation (BP) learning algorithm were used. This network was called Feed Forward Back-Propagation algorithm (FFBP), which consists of one or several hidden layer and one output layer [8-9]. For learning this network, BP learning algorithm is usually used. In the case of BP algorithm, the first output layer weight were updated. A desired value exists for each neuron of output layer. The weight coefficient was updated by the weight values and learning rules. During training this network, calculation were carried out from input of network toward output and values of error were then propagated to proior layers. Output calculations were conducted layer to layer so that the output of each layer was the input of next one. The Levenberg-Marquardt back propagation was used for updating network weights which, based algorithm for non-linear least squares optimization [8]. The steps of the training procedure are summarized as followed: (1) an input vector is applied, (2) the output of the network is calculated and compared to the corresponding targets vector, (3) the difference (error) is fed back through the network, and (4) weight are changed according to an algorithm that tends to minimize the error. The vectors of the training set are applied sequentially. This procedure is repeated over the entire training set for as many times as necessary until the outputs do not significantly change any more. After the end of training, simulation were done with the trained model to check the accuracy of the model using the determination coefficient (R^2) and calculated values for the models and root mean square error analysis (RMSE).

2.4 Designing the ANNs

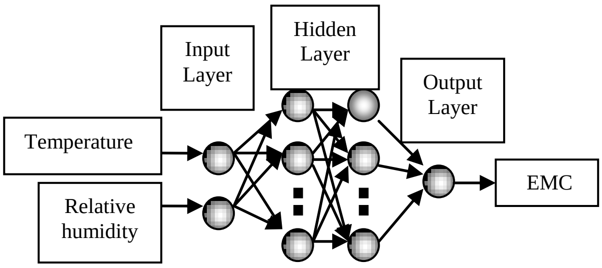

Fig 1. Artificial neural network topology

Considering and applying the two inputs in 80% of all experimental data to training data and residual data to testing data. Networks with two neurons in input layer are Relative humidity and temperature. One neuron in output layer (EMC) is design. Figure 1 showed the considered neural network topology and input and output parameters. The optimum neuron in the hidden layer obtains by adjustments and the transfer function were varied to reach the optimized.

2.5 Isosteric sorption heat [11]

The isosteric sorption heat is the required energy to remove water from the mass unit of a solid matrix. Hence, it is an assessment of the binding energy between water molecules and the solid matter of a food product [11]. The net isosteric heat of sorption phenomena can be explained by the Clausius-Clayperon equation as follows:

$$\frac{\partial \ln(RH)}{\partial T_{ab}} = \frac{\Delta H}{R_0 T_{ab}^{\;2}} \qquad (1)$$

Where RH is the relative humidity (%), T_{ab} is absolute temperature (K), △H is isosteric heat of sorption (kJ/mol) and R is universal gas constant.

Integrating equation (1) and assuming that the isosteric heat of sorption (△H) is independent of temperature, gives the following:

$$\ln(RH) = \left(\frac{\Delta H}{R_0}\right)\frac{1}{T_{ab}} + C \qquad (2)$$

The value of △H can be calculated from the slope of the equation.(2). From the thermodynamic relationship:

$$\Delta G = \Delta H - T_{ab}\Delta S \qquad (3)$$

Where △G is the Gibbs free energy (J/mol) and △s is the entropy.

For moisture sorption, it can be shown that:

$$\Delta G = -R_0 T_{ab} \ln(RH) \qquad (4)$$

Substitute △G from Eq. (4) into equation (2), the following equation is obtained:

$$\ln(RH) = \left(\frac{\Delta H}{R_0}\right)\frac{1}{T_{ab}} + \frac{\Delta S}{R_0} \qquad (5)$$

2.6 Drying equipment

The green house dryer showed in Fig. 2. The dimension of drying room was about 1×1.2×1.5 m^3. All sides of dryer were made of black plastic sheet for acting as solar collector whilst the translucent plastic sheet was used for making roof. At the bottom and top of dryer there were two inlet and two outlet air tubes for heating and exhausting air, respectively [15].

Fig. 3 showed the small scale drying unit with 20 rubber sheet samples capacity [15].

The drying temperature was measured by K-typed thermocouple connected to a data logger with an accuracy of ±1°C and was recorded every half an hour.

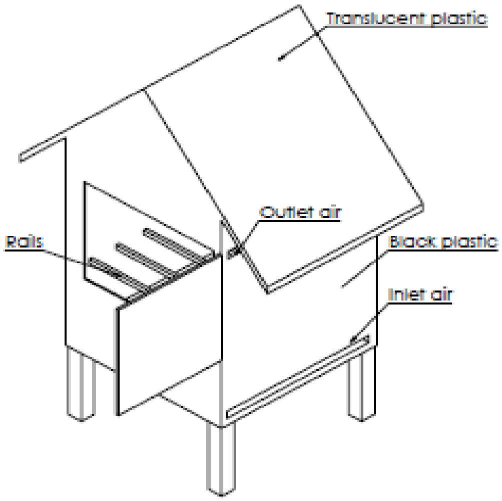

Fig. 2. Schematic diagram of green house drying system [15]

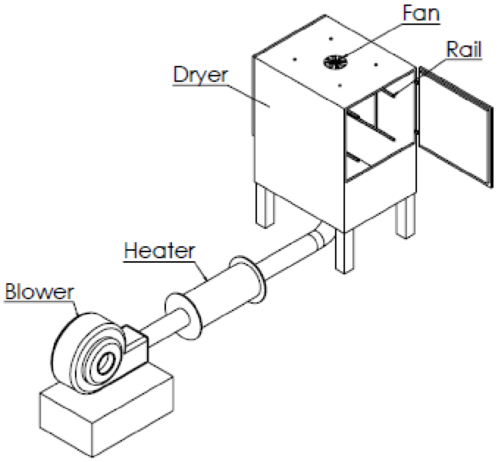

Fig. 3. Schematic diagram of hot air drying system [15]

2.7. *Drying kinetic and mathematics model*

Relationship between moisture ratios and drying time normally is determined by three major mathematical drying models as following: (1) simulation including heat and mass transfer, (2) the diffusion model and (3) the empirical or semi-empirical model mostly developed from experimental data. The first and second model also called as theoretical model and semi theoretical model.

The initial moisture contents of sample rubber were operated in range of 15 - 40 % dry basis and the desired final moisture content with all of was 1± 0.05% dry basis [2]. To simplify the predicted evolution of moisture content (MC) in this work, eventually the simplified equation can be noted as:

$$MC = \frac{W_i - W_d}{W_d} \times 100\% \tag{6}$$

Where MC is moisture content (% dry-basis), Wi is mass of rubber sheet at drying time (kg), Wd is mass of dry sample (kg).

The moisture ratio and drying rate of rubber sheet during drying experiments were calculated using the following equation:

$$\text{Moisture ratio} = MR = \frac{(M_t - M_g)}{(M_o - M_g)} \tag{7}$$

$$\text{Drying rate} = \frac{(M_i - M_f)}{\text{Drying time}} \times W_d \tag{8}$$

Where MR is moisture ratio (dimensionless), M_t is average moisture content at drying time (% dry-basis), M_i is initial moisture content (% dry-basis), M_e is equilibrium moisture content (% dry-basis), M_f is final moisture content of rubber sheet (% dry-basis), W_d is mass of dry sample (kg).

2.8. *Moisture diffusion coefficient*

Drying usually occurs in a number of stages with an initial constant rate period, followed by falling rate periods. In the constant rate period, the drying surface is saturated with water and drying rate is limited by the rate at which heat is transferred to the material. The falling rate periods indicate an increase in both heat and mass transfer resistances and they occur when moisture movement is controlled by internal mechanism.

Fick's second law of diffusion has been widely used to describe the drying process drying the falling rate period for the most biological material [3] can be express as

$$\frac{\partial M}{\partial t} = \nabla\left[D_{eff}(\nabla M)\right] \tag{9}$$

D_{eff} is the effective moisture diffusivity representing the conductive term of all moisture transfer mechanisms. This parameter is usually determined from experimental drying curves. Based on the assumptions of uniform initial temperature gradients, negligible external resistance, negligible temperature gradients, negligible shrinkage during drying and constant diffusion coefficient. From the thin layer drying and geometric shapes can be written in a differential equation of moisture diffusion that is the ratio of moisture to form slab sheets. So, the analytical solution of equation (9) for this case of an infinite slab is:

$$\mathrm{MR} = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\left[\frac{1}{(2n+1)^2}\right]\exp\left[\frac{-(2n+1)^2\pi^2 D_{eff}\,t}{L^2}\right] \tag{10}$$

Where L is small and t is large, the terms in the summation series of equation (10) corresponding to n>1 are small. Under these conditions the following approximation can be made:

$$\ln(MR) = \ln\left(\frac{8}{\pi^2}\right) - \frac{\pi^2 D_{eff} t}{L^2} \tag{11}$$

Where D is the effective diffusion coefficient (m2/h), t is the drying time (h), L is thickness of the slab sheet (0.0004185 m).

The effective diffusivity can be calculated from the slope of the plot ln(MR) vs time.

The effective moisture diffusivity for rubber sheet with increasing temperature. Temperature dependence of the effective diffusivity has been shown to follow an Arrhenius relationship [3]:

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{RT}\right) \tag{12}$$

Where D_0 is the Arrhenius factor of the heterogeneous solid (m^2/h), E_a is the activation energy (kJ/mol-K), R is the universal gas constant (8.314 kJ/mol-K), T is the absolute temperature (K).

The correlation coefficient (R) was primary criterion for selecting the best equation to describe the drying curve equation. The goodness of fit of each equation was evaluated using the determination coefficient (R^2) and calculated values for the models and root mean square error analysis (RMSE) were used to determine the goodness of the fit. The equations 13-14 were used for calculating R^2 and RMSE, respectively.

$$R^2 = \sqrt{1 - \frac{\sum_{i=1}^{n} (MR_i - MR_{pi})^2}{\sum_{i=1}^{n} (MR_i - MR_{mi})^2}} \qquad (13)$$

$$RMSE = \left[\frac{\sum_{i=1}^{N} (MR_{pi} - MR_i)^2}{N} \right]^{1/2} \qquad (14)$$

Where MR_i is the experimental value, MR_{pi} the predicted value, MR_{mi} the average of experimental values and N the number of observations. N is the number constants.

To analysis the mathematical modeling of thin layer drying equations of rubber sheet was predicted the evolution of moisture ratio against drying time. These empirical models and semi-theoretical model were develop from experimental results using the statistical nonlinear regression and the best fitting empirical model were showed in Table 1 [3,10,14].

Table 1. Different empirical model for rubber sheet drying

Model	**Equation No.**
Page MR = exp(-kt^n)	(14)
Two term exponential MR = a exp(-kt)+(1-a) exp (-kat)	(15)
Overhults MR= exp(-(kt)^n)	(16)
Approximation of diffusion MR = a exp(-kt)+(1-a)exp(-kbt)	(17)
Verma et al. MR = a exp(-kt)+(1-a)exp(-gt)	(18)
Midilli et al. MR = a exp(-ktn)+bt	(19)
Two term MR = a exp(-k_1t)+b exp(-k_2t)	(20)
Modified Henderson and Pabis MR = a exp(-kt)+b exp(-gt)+c exp(-ht)	(21)
Modified Page MR= aexp(-(kt)^n)	(22)
Weibull Distribution MR= a-bexp(-(kt^n))	(23)
C.L.Hii et al. MR = aexp(-kt^n)+b exp(-kt^n)	(24)
Modified Overhults MR = aexp(-(kt)^n)+b	(25)

Where MR is the moisture content (%dry-basis), t is drying time (h), and a, b, g, h, k, k_1, k_2, and n are the constant value.

3. Results and Discussion

3.1. Neural network model approach

The ANN model of equilibrium moisture content of natural was developed by training with experimental data at temperature 40, 50 and 60 °C and relative humidity of 10-90%. The isotherm data at 55 °C were reserved for testing the model. After 10 times iteration [7] step of training, the determination coefficient (R^2) and root mean square error analysis (RMSE) using to accuracy the network. Several topologies were tested and the best results which used from each from network, training algorithm and transfer functions are presented in Table 2.

Table 2. Training algorithm for different neurons and hidden layer for several network

Transfer function	No.of layer and neuron	R^2	RMSE	Epoch
tansig-tansig-tansig	2-3-3-1	0.9764	0.01426	16
	2-4-2-1	0.985	0.01736	16
	2-2-2-1	0.9764	0.02214	8
logsig-logsig-logsig	2-3-3-1	0.9958	0.01902	9
	2-4-2-1	0.9697	0.02674	7
	2-2-2-1	0.9836	0.02450	14
tansig-logsig-tansig	2-3-3-1	0.9805	0.02363	8
	2-4-2-1	0.9939	0.01408	5
	2-2-2-1	0.9752	0.02243	30
tansig-tansig-purelin	2-3-3-1	0.9718	0.02529	22
	2-4-2-1	0.9973	0.00731	16
	2-2-2-1	0.9588	0.02880	5
logsig-logsig-purelin	2-3-3-1	0.9846	0.01781	18
	2-4-2-1	0.9795	0.02291	8
	2-2-2-1	0.9684	0.02490	4
tansig-logsig-purelin	2-3-3-1	0.9898	0.02281	5
	2-4-2-1	0.9924	0.01787	6
	2-2-2-1	0.9694	0.02697	7

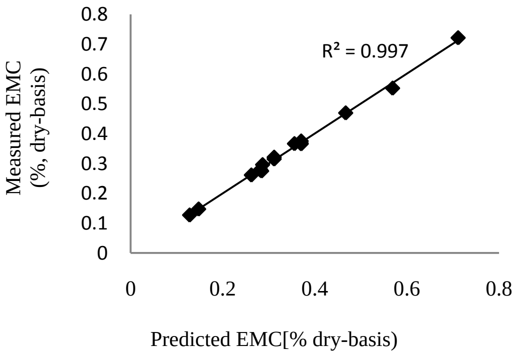

Fig.4. Predicted values of EMC using ANNs versus experimental values for training set at 40, 50 and 60 °C

The best results for FFBP network with LM algorithm (trainlm learning function) is tansig-tansig-purelin transfer function and 2-4-2-1 topology. The composition between experiment and predicted given R^2 =0.9973, RMSE = 0.00731 and converged in 16 epochs was showed in Fig. 4.

3.2. Isosteric heat of sorption

Equilibrium moisture content values of the unsmoked rubber sheet at the four temperature level were 40°C, 50°C, 55°C, and 60°C and eight equilibrium moisture content levels were 0.15%, 0.20%, 0.25%, 0.30%, 0.35%, 0.40%, 0.45%, 0.50%, which the value of ln(RH) versus 1/T were plotted at constant moisture content. The slope of the line was the net isosteric heat of sorption. The heat and entropy of sorption was found the to decrease with increasing moisture content can also seen in Fig. 5and6.

The net isosteric heat of sorption was found to fit a exponential relation with the R^2 values and RMSE values was 0.9712 and 0.1846 respectively. The following equation was developed for the unsmoked sheet

$$\Delta H = 514.43 e^{-7.571EMC} \tag{26}$$

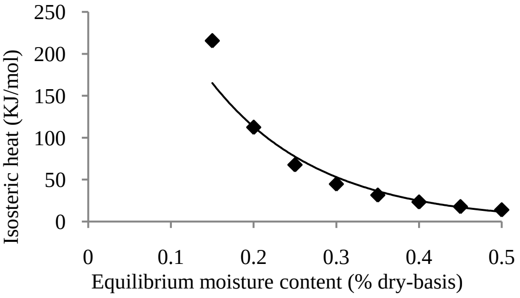

Fig. 5. heat of unsmoked rubber sorption at the different equilibrium moisture content

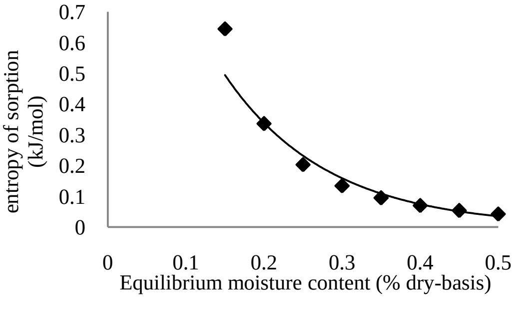

Fig. 6. entropy of unsmoked rubber sorption at the different equilibrium moisture content

3.3. Drying kinetic and mathematic model

The comparison of moisture ratio between experimental and predicted value of HA at drying temperature 45°C and 55 °C. Fig. 7 showed that the propose Weibull Distribution model could predict the moisture ratio in good agreement with the experimental results.

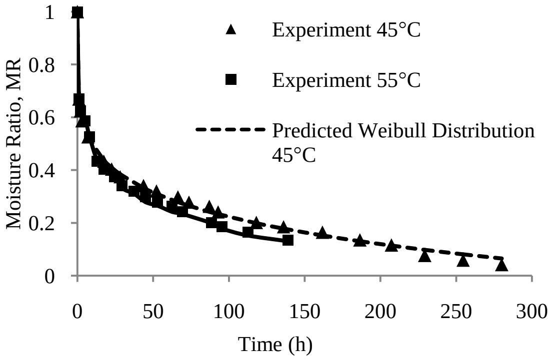

Fig. 7. Drying curves of HA drying (Weibull Distribution model) experimental and predicted, using the different air temperature values

The best model describing the thin layer drying characteristic was chosen as the Weibull Distribution model with the highest R^2 values were 0.99710 and 0.9955, respectively, the lowest RMSE values were 0.0125 and 0.01611, respectively.

Modified Henderson and Pabis could best fitting model for described OA and GH drying behavior of rubber sheet where R^2=0.9989 and R^2=0.9996, respectively showed in Fig. 8 and 9

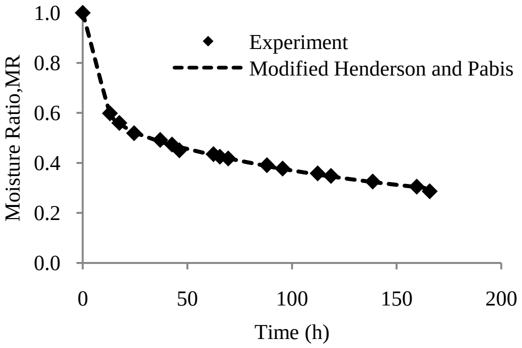

Fig.. 8. Drying curves of OA drying experimental and predicted

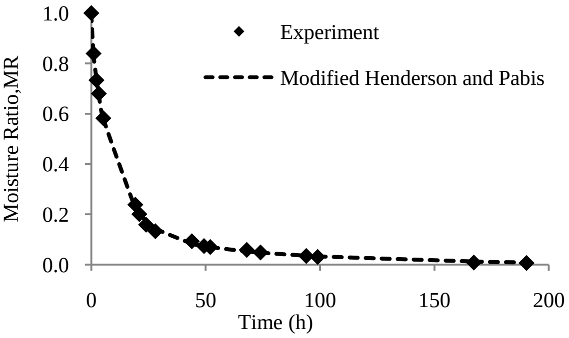

Fig. 9. Drying curves of GH drying experimental and predicted

At the beginning of drying time, the moisture ratio of sample rapidly decreased since the main part of moisture content of sample existed around the exterior surface, thus allowing the easier water removal without any interference of disordered void spaces inside sample. At nearly end of drying period, heat and mass transfer did not only occur at the surface of rubber sheet but also they stimulated inside rubber sheet. Nevertheless, the moisture inside rubber sheet moved to surface slower than the movement from the surface of rubber sheet to ambient environment. Drying rate will be relative lowers compared to the beginning of drying time. Additionally, at a higher drying air temperature, rate of moisture removal became relatively faster than those of a lower temperature.

In this study twelve models were used to fit the experimental data on moisture ratio and drying time using equation (14)-(25). Estimated parameters and the accuracy of the fit of different models was evaluated by calculating coefficient of determination (R^2) and root mean square error (RMSE), between the experimental data and predicted value for selected models of the best fitting of rubber sheet drying. The best fitting of rubber sheet drying of HA 45°C, HA 55°C, GH and OA are presented in Table 3

Table 3. Results of statistical analysis on the modeling of rubber sheet drying with HA temperature of 45°C, 55°C, GH and OA

Source	Model	Parameter	R^2	RMSE
HA 45°C	Weibull distribution	a = -2.35729	0.9971	0.0125
		b = -3.35363		
		k = 0.04256		
		n = 0.20900		
HA 55°C	Weibull distribution	a = -2.64859	0.9955	0.0125
		b = -3.64371		
		k = 0.02350		
		n = 0.28062		
OA	Modified Henderson and Pabis	k = 0.00016	0.9989	0.0051
		a = 0.30496		
		b = 0.28121		
		c = 0.41387		
		g = 1.8E-05		
		h = 0.00268		
GH	Modified Henderson and Pabis	k = 0.00024	0.9996	0.0064
		a = 0.13675		
		b = 0.11326		
		c = 0.7501		
		g = 0.02758		
		h = 0.00159		

3.4. Effect of temperature of rubber sheet

The sample was dried by HA with constant temperature of 45 °C and 55 °C, initial moisture content 17.86% dry-basis and 15.09% dry-basis, respectively. Fig. 10 showed that at the hot air drying temperature of 55 °C, the reduction of moisture was faster than that of a hot air dying temperature of 45 °C. Furthermore, in the beginning of drying time, the moisture ratio of sample rapidly increased, which moisture ratio decreased with the increasing of drying time

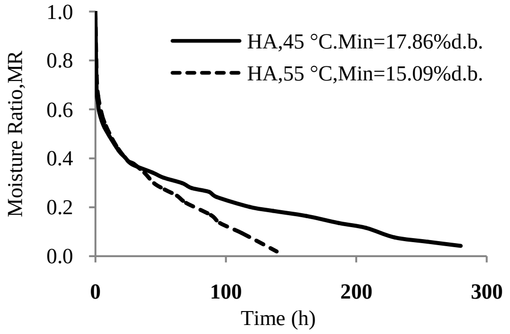

Fig. 10. Effect of temperature of rubber sheet at drying temperature of 45 °C and 55 °C of HA (initial moisture contents of 15.09-17.896 % dry-basis)

The rubber sheet was dried by natural sun drying took longer period than those of drying. (Fig. 11)

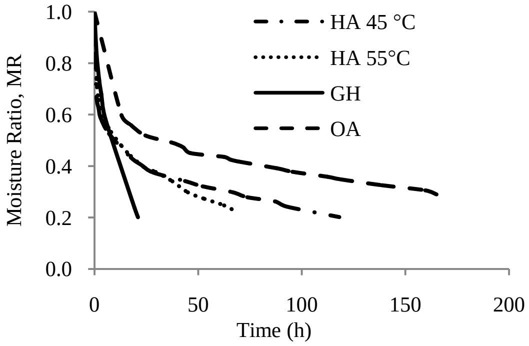

Fig. 11. Comparison of drying time of rubber at 30-55 °C at relative humidity 50-60%dry-basis

3.5. Moisture diffusion coefficient of rubber

Experiment drying a thin layer of rubber was dried in the drying rate decreased. The moisture diffusion coefficient of the rubber can be calculated by equation (11) the results of moisture diffusion coefficient of rubber showed in Table 4:

When calculating the moisture diffusion coefficient, it was found that the moisture diffusion coefficient of the rubber decreased with increasing time. HA of temperature 45 °C took longer period drying than HA of temperature 55 °C.

It can also seen that HA of temperature 45 °C had the less moisture diffusion coefficient than HA of temperature 55 °C.

Table .4. Results of effective moisture diffusivity of rubber sheet drying with HA temperature of 45°C, 55°C, GH and OA

source	Diffusivity	source	Diffusivity
HA 45°C	1.5616x10-8	GH	4.5784x10-8
HA 55°C	3.6911x10-8	OA	9.2277x10-9

4. Conclusions

Main conclusion drawn from the results of the present study may be listed as follow:

1) The ANN model with two inputs (temperature and relative humidity), one output (EMC) and two hidden layers was found to be able to predict the EMC. The ANN model using transfer function of tansig-tansig-purelin and 2-4-2-1 topology given R^2 =0.9973, RMSE = 0.00731 found to be the good agreement between experiment and predicted values. The EMC of natural rubber could be predicted by ANN method.

2) The heat and entropy of sorption was found the to decrease with increasing moisture content

3) The Weibull Distribution were the best fitting model for describing HA drying (at different temperature of 45°C, 55°C), and the Modified Henderson and Pabis had a good relation to the experimental values of sample drying with GH drying and OA drying. It was found that drying kinetic providing the highest R^2 were 0.9971, 0.9955, 0.9996 and 0.9989, respectively and the lowest RMSE were 0.0125, 0.0125, 0.0064 and 0.0051, respectively. Both model could predict the moisture ratio in good agreement with the experimental results. Furthermore, quality of the rubber sheet was acceptable in market level.

4) At the beginning of drying time, the moisture ratio of sample rapidly increased, moisture ratio decreased with the increasing of drying time

5) The rubber sheet was dried by OA drying took longer period than those of drying.

6) The moisture diffusion coefficient of the rubber decreased with increasing time.

7) The quality of the rubber sheet was acceptable in market level. (Standardize ADS quality)

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