

## Mathematical Modelling of Sun Drying Kinetics of Thin Layer Cocoa (*Theobroma Cacao*) Beans

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**Abstract:** In this work, the sun drying behaviour of cocoa beans was investigated. Drying experiments were conducted for organic fermented cocoa beans grown in Yamoussoukro, Ivory Coast. The experimental drying curves obtained show only a falling rate period. In order to estimate and select the suitable form air drying curves, the drying data were fitted to fourteen different mathematical models. Coefficient of determination ( $R^2$ ) and other statistical parameters, such as reduced chi-square ( $x^2$ ), root mean square error (RMSE), mean bias error (MBE) and Student t statistic were used for determination of the best suitable model. Among the mathematical models investigated, the logarithmic model satisfactorily described the drying behaviour of cocoa beans with highest  $R^2$  and lowest  $x^2$ , RMSE and MBE. The apparent diffusion coefficient, varying from  $3.70 \times 10^{-11}$  to  $5.80 \times 10^{-11}$  m<sup>2</sup>/s, is an important parameter in the moisture transfer, and they were found to be dependent on the temperature, relative humidity and velocity of the drying air. The activation energy was estimated to 22.48 KJ/mol.

**Key words:** Cocoa beans, Drying kinetics, Thin layer models, Diffusion coefficient, Activation energy.

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### INTRODUCTION

Cocoa beans are the seeds of *Theobroma cacao* (Sterculiaceae family), a tropical tree. It is grown mostly in the wet tropical forest climate countries such as Ivory Coast, Ghana, Nigeria, Brazil and Malaysia <sup>[1]</sup>. The three varieties are Forasteros, Trinitarios and Criollos; with Amelonado (of the Forasteros) being the most abundant, while Criollos has become negligible in world trade.

Cocoa is used in the production of chocolate bars, milk chocolate, cocoa powder, cosmetics and pharmaceutical products. Others include the use of the shells for stock feed and manure. It is also a source of theobromine, shell fat and vitamin D. The pod is also rich in potash and is used for soap production.

Then, after harvesting of ripe cocoa pods, fresh cocoa beans are fermented for 5-7 days and dried immediately after fermentation to safe moisture level of 7.5% (wet basis). During these stages the cocoa beans undergo various chemical and biochemical changes that form the necessary flavour precursors needed during processing. Drying is usually carried out using natural sun drying.

Drying is the most important process to preserve grains, crops and foods of all varieties. The removal of moisture prevents the growth and reproduction of microorganisms causing decay and minimises many of the moisture-mediated deterioration reactions. It brings about substantial reduction in weight and volume, minimising packing, storage and transportation costs and enables storability of the product under ambient temperatures <sup>[2]</sup>.

Drying is a complex thermal process in which unsteady heat and moisture transfer occur simultaneously <sup>[3]</sup>. The description and prediction of the drying kinetics of a given material, under given process conditions, is still a weakness in the modelling of drying processes. Even now, in design and optimization of drying processes, there is a great need for stable and reliable models to quantify and predict drying rates and drying times with a satisfying accuracy. Over the last decades several approaches have been proposed about how to deal with mass and heat transfer phenomena in materials during a drying process.

The models fall into three categories namely the theoretical, semi-theoretical and empirical. Semi-theoretical models offer a compromise between theory

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and ease of application [4]. Examples of semi-theoretical models are such as the Newton model [5], Page model [2], Henderson and Pabis model [6], logarithmic model [7], two-term model [8], two term exponential model [9], Verma et al. model [10] and the Midilli–Kucuk model [11] are used widely. In empirical modelling a direct relationship between the moisture content and drying time is derived. The fundamentals of the drying process are neglected and the Wang and Singh model is an example of empirical model used in literatures [12].

Recently, many authors have undertaken studies covering mathematical modelling and kinetics of the foodstuff drying process (e.g. red chilli [13], apple [14], amaranth grain [15], mulberry [16], okra [17] and tomato [18]).

Studies of the modelling of cocoa drying are relatively scarce and only few published literatures are available [19-22].

Most of the cocoa drying literatures are focused mostly in flavour development, quality and bean acidity [23-26].

The present study aimed to observe the effect of air characteristics on cocoa beans drying behaviour, evaluate a suitable drying model for describing the drying process and finally to compute effective moisture diffusivity and activation energy of samples.

### MATERIAL AND METHOD

**Drying Experiments:** The solar drying experiments were conducted during the period of May to August 2007 in Yamoussoukro (a city in the centre of Ivory Coast, West Africa).

Each experiment started at 8:30 a.m. and continued until 5:00 p.m. During the experiments, daily solar radiation changed between 34 and 1406 W/m<sup>2</sup>, ambient air temperature ranged from 29.66 to 31.66°C, ambient air relative humidity from 57.50 to 76.50%, and drying air flow rate from 0.76 to 1.21 m/s. The solar radiation energy was maximum at midday and minimum at evening in all the days of experiments.

Drying experiments were performed in a 1 m<sup>2</sup> wooden tray situated in an open weather station. The station includes anemometer, psychrometer and thermometers for air velocity, relative humidity and temperature respectively determination. The moisture losses were determined by a digital balance with accuracy 0.01g.

About 20 kg fermented cocoa beans supplied by an organic agricultural farm were spread on the tray. Drying of beans started with an initial moisture content around 55-60 % (by wet weight) and continued until no further changes in mass were observed, e.g. to the final moisture content of about 7.5 % (by wet weight) which was taken as the equilibrium moisture in later computation.

At the end of each drying experiment, the final moisture of the sample was determined. Moisture contents were reported as wet-basis percentages, using the mean final moisture content and weight of the dried beans to calculate the weight of dry solids. Then, the moisture contents were expressed as a dry basis, which is more convenient for modelling.

While air velocity, relative humidity and temperature are measured continuously, cocoa mass loss different measurements occurred with an interval of 30 min. All drying experiments were triplicated.

**Modelling of Drying Curves:** The experimental moisture content data of organic cocoa beans obtained were fitted to 14 thin layer drying models summarised in Table 1 by using non-linear least squares regression solved by a Levenberg-Marguardt numerical method. The dimensionless moisture ratio (MR) was simplified to  $M/M_0$  instead of  $(M-M_e)/(M_0-M_e)$  for long drying times because  $M_e$  is relatively small compared to  $M$  or  $M_0$  [16].

The purpose of the fitting is to find out the best suited model for describing the drying curve of cocoa beans. The linear and non-linear regression analysis was performed using Matlab R2007b software (MathWorks Inc., Massachusetts, USA). The coefficient of determination ( $R^2$ ) was the primary criterion for selecting the best model to describe the drying curves [7,37]. In addition to  $R^2$ , the statistical parameters, such as reduced chi-square ( $\chi^2$ ), root mean square error (RMSE), mean bias error (MBE) and Student t statistic were calculated to evaluate the fitting of a model to experimental data. The highest values of  $R^2$  and the lowest values of  $\chi^2$ , RMSE, MBE and t-values were used to determine the best fit [9,11,38,39]. These statistical parameters were calculated as follows:

$$\chi^2 = \frac{\sum_{i=1}^N (MR_{exp,i} - MR_{pred,i})^2}{N - n} \quad (1)$$

$$RMSE = \left[ \frac{1}{N} \sum_{i=1}^N (MRE_{exp,i} - MRE_{pred,i})^2 \right]^{1/2} \quad (2)$$

$$MBE = \frac{1}{N} \sum_{i=1}^N (MR_{pred,i} - MR_{exp,i}) \quad (3)$$

where  $MR_{exp,i}$  is the  $i_{th}$  experimentally observed ratio,  $MR_{pred,i}$  the  $i_{th}$  predicted moisture ratio,  $N$  the number of observations and  $n$  is the number of constants [7]. Stone [40] reported that the MBE and RMSE do not represent reliable assessment of the model performance. To avoid false selection of the best model, t-value (Equation 4) was considered which must be smaller than the value for that confidence level in standard statistical tables.

**Table 1:** Mathematical models given by various authors for the drying curves

Model no	Model equation	Name	Reference
1	$MR = \exp(-kt)$	Newton	[5]
2	$MR = \exp(-kt^n)$	Page	[27]
3	$MR = \exp(-kt^n)$	Modified Page I	[28]
4	$MR = a \exp(-kt)$	Henderson and Pabis	[29]
5	$MR = a \exp(-kt+c)$	Logarithmique	[30]
6	$MR = \exp(-k_0t) + b \exp(-k_1t)$	Two-term	[31]
7	$MR = 1+at+bt^2$	Wang and Singh	[12]
8	$t = a \ln MR + b (\ln MR)$	Thomson	[32]
9	$MR = a \exp(-kt) + (1-a) \exp(-kbt)$	Diffusion Approach	[33]
10	$MR = a \exp(-kt) + (1-a) \exp(-gt)$	Verma	[10]
11	$MR = a \exp(-kt) + b \exp(-gt) + c \exp(-ht)$	Modified Henderson Pabis	[34]
12	$MR = a \exp(-kt) + (1-a) \exp(-kbt)$	Two term exponential	[35]
13	$MR = a \exp(-c(t/L^2))$	Simplified Fick's diffusion	[36]
14	$MR = \exp(-k(t/L^2)^n)$	Modified Page equation II	[36]

$$t\text{-value} = \sqrt{\frac{(n-1)MBE^2}{RMSE^2 - MBE^2}} \quad (4)$$

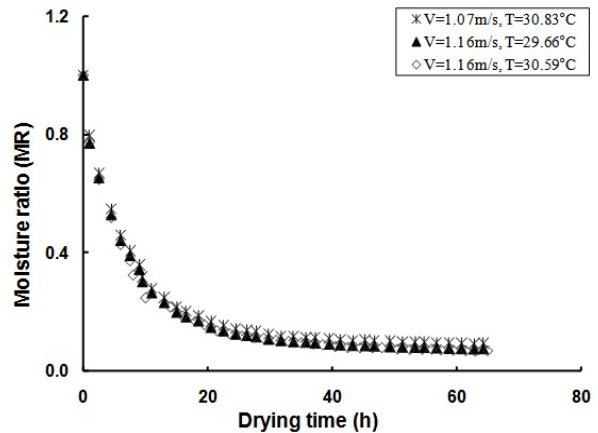
**RESULTS AND DISCUSSION**

**Drying kinetics:** The cocoa beans, considered as single layer, were dried at open sun from an initial moisture content of around 122 kg water per kg dry matter to the final moisture content of about 8 kg water per kg dry matter until no further changes in their mass were observed. This final moisture content has represented the moisture equilibrium between the cocoa beans and drying air under the experimental conditions.

Moisture ratio versus drying time is given in figure 1. It was observed that moisture ratio decreases continuously with drying time and no constant drying rate period exists. The drying rates were observed in the falling rate period in all drying conditions. Higher drying time (about 65 hours) was required to remove the moisture content and it may be due to the slow diffusion process. These results are in agreement with previous observations of different foods drying [2,9,39, 41,42].

**Modelling of Drying Curves:** The moisture content data obtained during drying experiments were converted into moisture ratio expression and then curve fitting computations with drying time were performed for all

fourteen drying models given in table 1. It was observed from the statistical analysis results (table 2), that high determination coefficients ( $R^2 > 0.85$ ) are found for the drying models except diffusion approach ( $R^2 = 0.630$ ) and two terms exponential ( $R^2 = 0.646$ ) models. The results showed that the highest values of determination coefficient were obtained with logarithmic and Verma models. However, logarithmic model gives lower MBE, RMSE,  $\chi^2$  and t-value than that of Verma model. Therefore, logarithmic model may be assumed to represent the thin-layer drying behaviour of fermented cocoa beans.



**Fig. 1:** Variation of moisture ratio with drying time of cocoa beans for different drying air conditions

In order to account for the effect of the drying variables on the logarithmic model constants a, k and c were regressed against those of drying air temperature, relative humidity and velocity using multiple regression analysis. Then, the obtained model for the moisture ratio is as follows:

$$MR = \frac{M}{M_0} = a \exp(-kt) + c$$

with :

$$\left. \begin{aligned} a &= 0,8969 + 0,0021T - 0,0917V & r &= 0,930 \\ k &= -0,1576 + 0,0065T - 0,0006HR + 0,1272V & r &= 0,909 \\ c &= 0,2015 - 0,0024T - 0,0019HR + 0,0866V & r &= 0,840 \end{aligned} \right\} (5)$$

where a and c are dimensionless coefficients and k (s<sup>-1</sup>) is the drying constant.

These expressions can be used to estimate the moisture ratio of cocoa beans at any time during the open sun drying with a good accuracy.

Validation of this selected model was confirmed by comparing the predicted moisture contents with the measured ones. Indeed, the plot presented in figure 2 shows slight discrepancy between both values because the data are closely scattered around the straight line. Therefore, a good agreement between experimental and predicted moisture ratio values is observed with a R<sup>2</sup> superior to 0.976.

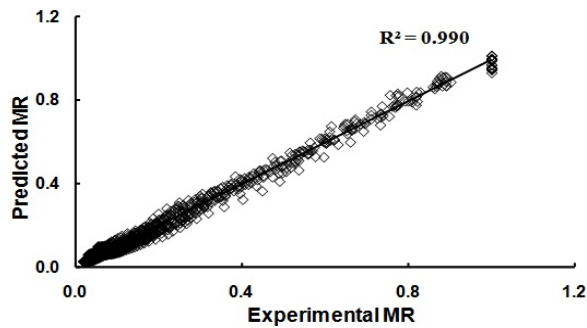


Fig. 2: Comparison of experimental and predicted values moisture ratio (MR) by the logarithmic model for different drying conditions

**Determination of the Effective Moisture Diffusivity:**

Fick's second law can be used to describe the drying behaviour. The solution of Fick's second law in slab geometry, with the assumptions of moisture migration being by diffusion, unidimensional moisture movement, negligible shrinkage, constant diffusion coefficients and temperature was as follows [43]:

$$MR = \frac{M}{M_0} = \frac{8}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} \exp\left(-\frac{(2n-1)^2 \pi^2 D_{eff} t}{4L^2}\right)$$

where D<sub>eff</sub> is the effective diffusivity coefficient (m<sup>2</sup>/s); L is the half thickness of the slab (m) and n is the positive integer.

For long drying period the above equation can be simplified, by taking the natural logarithm of both sides:

$$\ln MR = \ln \frac{8}{\pi^2} - \left(\frac{\pi^2 D_{eff} t}{4L^2}\right)$$

From the logarithmic drying curves, the D<sub>eff</sub> values were obtained at different drying conditions. The results are presented in figure 3. It appears that diffusion coefficient, very low at drying starting (0.001x10<sup>-11</sup>m<sup>2</sup>/s), increases till a maximum value of 8.76x10<sup>-11</sup> m<sup>2</sup>/s after 10 hours of drying, corresponding to a moisture ratio of 0.35. After this time, the diffusion coefficient decreases to about 2.45-4.35x10<sup>-11</sup> m<sup>2</sup>/s, according to the different drying conditions. The mean diffusion coefficient values obtained vary therefore in the interval 3.78-5.38x10<sup>-11</sup> m<sup>2</sup>/s.

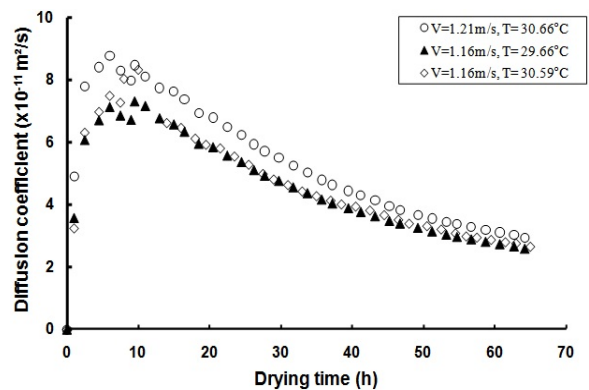


Fig. 3: Variation of diffusivity D<sub>eff</sub> with drying time at different temperatures and velocities

**Activation Energy:** The effective diffusivity coefficient depends on temperature and can be described by Arrhenius equation [44]:

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

where D<sub>0</sub> is the constant in Arrhenius equation (m<sup>2</sup>/s); E<sub>a</sub> is the activation energy (kJ/mol); T is the drying air temperature (K) and R is the universal gas constant (kJ/mol.K). Activation energy for diffusion was estimated by using this equation.

This equation can be linearised by applying natural log at both sides and a plot of ln (D<sub>eff</sub>) versus 1/T will produce a straight line.

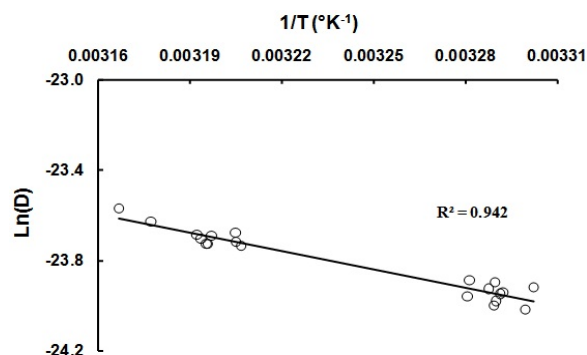
**Table 2:** Values of the drying constants and coefficients of mathematical models determined through regression method for cocoa beans

Model n°	Name	Model constant	R <sup>2</sup>	MBE	RMSE	c <sup>2</sup>	t- value
1	Newton	k = 0.0968	0.972	-0.0366	0.0608	3.456 E-3	0
2	Page	k = 0.2334 n = 0.6564	0.974	-0.0189	0.0245	0.566 E-3	1.2099
3	Modified Page	k = 0.1065 n = 0.6564	0.974	-0.0154	0.0245	0.566 E-3	0.8062
4	Henderson and Pabis	a = 0.7834 k = 0.0697	0.938	-0.0276	0.0446	1.869 E-3	0.7871
5	Logarithmic	a = 0.8676 k = 0.1230 c = 0.0837	0.976	-0.0051	0.0128	0.144 E-3	0.6173
6	Two term	a = 0.4318 k <sub>0</sub> = 0.0797 b = 0.4318 k <sub>1</sub> = 0.0797	0.954	-0.0316	0.0519	2.500 E-3	0.7672
7	Wang and Singh	a = -0.0508 b = 0.0006	0.857	0.0359	0.1265	14.496 E-3	0.2965
8	Thomson	a = 0.6114 b = 7.8658	0.940	0.6170	3.0058	8146.26 E-3	0.2098
9	Diffusion approach	a = -0.0001 k = -19.949 b = -8.6036	0.63	0.1270	0.2899	80.647 E-3	0.6889
10	Verma	a = 0.2085 k = 0.0195 g = 0.1618	0.976	-0.0085	0.0146	0.197 E-3	1.0054
11	Modified Henderson and Pabis	a = 0.2879 k = 0.0797 b = 0.2879 g = 0.0797 c = 0.2879 h = 0.0797	0.954	-0.0315	0.0519	2.51 E-3	1.7155
12	Two term exponential	a = -2.4016 k = -0.0022	0.646	0.1192	0.2797	74.807 E-3	0.4709
13	Simplified Fick's diffusion (SFFD) equation	a = 0.8637 c = 1132.3160 L = 118.0535	0.958	-0.0347	0.0519	2.495 E-3	-1.2735
14	Modified Page equation II	k = 0.0102 n = 0.6637 L = 0.0961	0.974	-0.0197	0.0274	0.677 E-3	1.4719

A plot of the herein data is as shown in figure 4 and confirmed the proposed relationship between the effective diffusivities and temperatures ( $R^2 = 0.943$ ). The activation energy and the Arrhenius constant, that is a diffusivity constant equivalent to the diffusivity at infinitely high temperature, can be determined from the slope and the y-intercept, respectively. The activation energy is the energy barrier that must be overcome in order to activate moisture diffusion. By increasing the temperature and hence the drying rate this energy barrier can be overcome easier relatively but there should be a compromise between high temperature and

acceptable product quality [45]. Too high temperature is not recommended for cocoa drying as the resulting product is often associated with high acidity [24].

The values of  $D_0$  and  $E_a$  were estimated at  $2.91 \times 10^{-7} \text{ m}^2/\text{s}$  and 22.48 KJ/mol, respectively. These values are actually within the range for other agriculture materials [4,41]. But the results published by Hii *et al.*, [19] show higher values diffusivities than those presented herein. The differences observed are related to higher drying temperatures (i.e. 40, 60, 70°C) used in their study.



**Fig. 4:** The Arrhenius relationship between diffusivities and temperature

**Conclusion:** The logarithmic drying model, which gave a higher value for the coefficient of determination and lower values for the root mean square error, mean relative percent error, reduced chi-square and t-value among the 14 models was considered the best model for describing the drying behaviour of organic cocoa beans. Also, the established model developed for describing the drying behaviour of organic cocoa beans as affected by the drying air temperature, relative humidity and velocity was found to fit the experimental data reasonably well based on statistical analysis.

The apparent diffusion coefficient, varying from  $3.70 \times 10^{-11}$  to  $5.80 \times 10^{-11}$  m<sup>2</sup>/s, is an important parameter in the moisture transfer, and they were found to be dependent on the temperature, relative humidity and velocity of the drying air.

The activation energy was found as 22.48 KJ/mol.

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