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Modelling of experimental drying kinetics of Codiaeum Variegatum Brilliantissima-Zanzibar used as a corrosion inhibitor

Izionworu Onuegbu Vincent[1,*], Ojong E. Ojong[2], Ademiluyi Falilat Taiwo[1], Mohammad Fakhratul Ridwan Zulkifli[3], Walid Daoudi[4], and Wan Mohd Norsani Wan Nik[3]

[1]Department of Chemical/Petrochemical Engineering, Faculty of Engineering, Rivers State University, Port Harcourt, Nigeria
[2]Department of Chemical Engineering, Faculty of Engineering &Technology, University of Calabar, Calabar, Nigeria
[3]Faculty of Ocean Engineering Technology, Universiti Malaysia Terengganu, Terengganu, Malaysia
[4]Laboratory of Materials and Environment, Department of Chemistry, Multidisciplinary Faculty of Nador, University of Mohamed 1, Nador, Morocco
[*]Corresponding author: vincent.izionworu@ust.edu.ng

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Abstract

The drying kinetics of Codiaeum Variegatum Brilliantissima-Zanzibar (Wire Croton) in the open air were modelled in this research. Daily temperature and the humidity of the environment were monitored every 12 hrs during the experiment. Wire Croton drying results obtained every 12 hrs were plotted against drying time to study the drying properties of the sample at 12 and 24-hr intervals. The results obtained were fitted into Lewis, Page, Modified Page, Handerson-Pabis and Ademiluyi's model for the determination of the Root Mean Square Error (RMSE), which gave values of 0.277, 0.115, 0.173, 0.163 and 0.170, respectively and the coefficient of regression (R^2) values of 0.539, 0.525, 0.954, 0.539 and 0.954, respectively for 12 hourly test. The 24-hour interval test gave RMSE values of 0.218, 0.113, 0.101, 0.162 and 0.097, corresponding R^2 values of 0.801, 0.448, 0.448, 0.801 and 0.851. The Izionworu & Ojong model was developed and tested. When compared with existing models, Izionworu & Ojong's thin-layer model gives the least RMSE value of 0.104 and 0.029 at 12 and 24-hour intervals respectively, compared to the other models and an R^2 of 0.988 and 0.918 correspondingly for both the 12 and 24 hrs drying intervals. A 50% error deviation was achieved when the Izionworu & Ojong model was compared with the other two-term models confirming that the drying characteristics of wire croton in the open air are best described by the Izionworu & Ojong model.

Keywords: Codiaeum Variegatum Brilliantissima-Zanzibar, Drying kinetics, Model, Moisture content

1. Introduction

Codiaeum Variegatum Brilliantissima, also known as Wire Croton (WC) (fire croton, garden croton, or variegated croton; syn. *Croton variegatum* L.), is a species of plant in the genus Codiaeum, which is a member of the Family Euphorbiaceae. It is commonly found in Indonesia, Malaysia, Australia, and the western Pacific Ocean islands as a native plant that grows in the open forests and scrub. This plant has also been found in Rivers State University, Nkpolu–Oroworukwo, Port Harcourt, in Rivers State, Nigeria. The specie under review has leaves that are between 20-30 cm in length and about 7-10 mm broad.

The search for alternative sources of inhibitors for acid corrosion prevention leads to the use of this plant's extract as an inhibitor in the control of mild steel corrosion control in an acidic environment [1]. In practice, plant extract corrosion inhibitors are a cost-effective and easy-to-application method to control corrosion in acid and seawater media. The industrial practice is to apply organic and inorganic inhibitors to replace the toxic inorganic inhibitors, such as phosphates, chromates, nitrates, etc [2]. Some definite classes of organic and inorganic inhibitors have been found to be more environmentally susceptible: plant extracts, drugs, and rare earth compounds. Plant extracts such as *Moringa oleifera*, *Andrographis paniculate*, *Theobroma Cocoa* pod, *Piper Bitle*, and *Leucaena leucocephala* leaves have been researched as environmentally friendly corrosion inhibitors [3-7]. Although recent research has among other things focused on carbon dot [8] grafted chitosan [9,10] and isatin [11], Codiaeum Variegatum Brilliantissima is of current interest as it has successfully inhibited mild steel corrosion in hydrochloric acid environment.

Drying, which is a unit operation [12], is employed in the processing of WC. As a physical process, drying is characterized by the evaporative removal of water in solids. Drying is far less expensive as a means of moisture removal due to its low-cost heating source [13]. Drying is used to eliminate moisture from the WC to enable longtime storage and its aqueous extract to be used as a green inhibitor. Models have been developed to achieve large-scale drying of leaves, seeds, etc [14]. The drying characteristic of materials – leaves and seeds has been expressed as either a drying curve or drying rate curve [15] by different researchers using models like Page model [16], Newton model [15] and Henderson and Pabis model [17]. Some have also used the logarithmic model [16] and two-term exponential models [14].

Some examples of these are the moisture removal properties of red pepper, potato, and tomato when subjected to forced moisture removal, which is best described by the page model [13, 14, 18, 19]. In contrast, the exponential model has been found to describe the drying behavior of mulberry under the open sun [13,19] and the Newton model when strawberry is dried using solar [20], drying fermented cassava in a rotary dryer [21] and drying of different varieties of popcorn [22]. There is, however, no research literature on the natural drying of WC in a shade, that is, open-air drying or forced convection [23]. Aremu and Akintola [12], after citing several drying processes used by different researchers, summarized that the drying of the materials took place under a falling rate period, with the drying temperature being the most important parameter.

Drying under solar or sunlight has been adopted as a local drying technique [16, 20, 23]. This results in a slow weight drop during drying with some major setbacks, which include an extended drying time, the need for large areas with possible contamination with unwanted materials and, worse of all, loss of phytochemical components of the plants [16, 20] with possible reduced corrosion inhibition effect. Some researchers have dried plants and plant yields such as pepper and corn under temperatures higher than ambient temperatures [21, 22]. Others dry fresh leaves under shade to preserve their components but with the disadvantage of longer drying time. The preservation of the phytochemical components of leaves or plant extracts have been demonstrated as key to their use as corrosion inhibitors by several researchers [1-4]. Like other plants, the use of WC as a corrosion inhibitor requires the retention of its bioactive components. This draws attention to open-air drying as a good option to dry leaves in temperate regions of the earth with poor and epileptic electrical power supply.

Thin-layer drying models have not been applied to WC in the open air. So it is the purpose of this study to (i) apply five known thin-layer drying models and a newly-proposed thin-layer drying model to WC drying in the open air and at ambient temperature, (ii) demonstrate the efficiency of drying WC in the open air in the shade in tropical regions of sub – Sahara Africa using the Niger Delta region of Nigeria as a case study to save the cost of processing WC to be used as a corrosion inhibitor, and (iii) identify the limitations of the open air ambient drying as a drying option for preservation of the bioactive compounds and the functional groups in WC used as corrosion inhibitor for metals.

2. Materials and methods

2.1 Materials

The WC used was sourced from the garden in Chemical & Petrochemical Engineering Laboratory at Rivers State University (RSU). Stainless steel trays and wall clock. max-min indoor thermo and hydro with model number TA218A/B were used to record temperature and relative humidity measurements just above the WC leaves. The device measuring temperature range is: indoor 0°C~50°C(32°F~+122°F). Resolution: temperature 0.1°C; accuracy: ±1°C; Humidity 10%RH ~ 98%rh; Resolution: 1%RH, and Accuracy: 60% ±5%.

2.2 Methods

The WC leaves sample was first cut from the branch, washed, and cleaned of dirt and other impurities that could affect the experimental result. 600 g of fresh WC leaves were weighed out of the collected sample, and each leaf was cut into 1 x 10 cm. The cut 600 g of WC was divided into three portions, each weighing 200 g. Each portion of the 200 g of WC was spread on one stainless steel and left to dry in the open air in the RSU Chemical Engineering Laboratory. The three samples were to ensure reproducibility. Weighing and drying every twelve hrs intervals was done until weight loss became negligible. It was then dried in the oven overnight to get the dry bone weight at the point where the equilibrium moisture content was zero. The laboratory ambient temperature and humidity were recorded and reported in Table 1. This data gave an insight into the temperature and humidity of the environment during the drying of WC in a tropical region, and its dry matter weight allowed the construction of the sample drying curve.

Table 1 Air temperature and humidity readings.

Day	Temperature (°C)			Humidity (%)		
	Morning	Afternoon	Evening	Morning	Afternoon	Evening
1	28.37	30.98	33.27	70	58	53
2	28.60	30.40	30.10	68	62	61
3	29.40	31.90	31.00	67	59	62
4	28.0	31.30	31.81	71	67	62
5	28.40	29.90	30.50	73	70	66
6	28.10	32.10	33.00	73	62	58
7	29.10	28.00	28.60	68	72	73
8	30.00	32.60	32.50	71	62	62
9	29.40	31.00	29.80	77	68	74
10	29.9	31.60	31.60	74	66	66
11	30.20	32.30	32.31	68	71	68

2.2 Mathematical model formulation

Drying models from the first principle was considered using a thin layer drying model to determine the mathematical model to study the drying behavior of WC, taking into consideration the controlling mechanisms. In thin-layer drying, the moisture ratio during drying is calculated according to Equation (1),

$$MR = \frac{M\text{-}M_e}{M_o\text{-}M_e} \quad (1)$$

where M is the average moisture content at time t, MR is the dimensionless moisture ratio, M_O and M_e are the initial moisture content and the equilibrium moisture content, respectively, on a dry weight basis.

Throughout the thin layer wire croton drying in the oven dryer, the sample was not exposed to uniform relative humidity and temperature continuously. The equilibrium moisture content was assumed to be zero. So, the MR (Equation (1)) was reduced [23] as seen in Equation (2):

$$MR = \frac{M}{M_o} \quad (2)$$

The drying curve plots were done using the recorded moisture loss for the sample using CurveEpert Basic 22.3 software. Mathematical models of Lewis, Page, Modified Page, Henderson & Pabis and Ademiluyi Model presented in Table 2 show the connection between MR and the time of drying, t was applied to give the resulting drying curves achieved for WC at the test temperature and non-linear regression analysis was used to select the model that best describes the drying characteristics. Origin 8 computer software package was used to carry out regression analysis. Although different statistical parameters can be used to verify the model that best describes the drying of a vegetable, the correlation coefficient (R^2) and root mean square error (RMSE) were used in this research as the major criteria in the selection of the best model equation to describe WC's drying curve in ambient condition in sub-Sahara Africa. For an acceptable fit, a higher R^2 value is desired, while the RMSE ought to be low [21, 22]. The mean relative error and RMSE [11] are calculated using:

$$X^2 = \frac{\sum_{i=1}^{N}(MRpre,i\text{-}MRexp,i)^2}{N\text{-}Z} \quad (3)$$

$$RMSE = \left[\frac{\sum_{i=1}^{N}(MRpre,i\text{-}MRexp,i)^2}{N\text{-}Z}\right]^{0.5} \quad (4)$$

where MRexp, i is the experimental moisture ratio and, MRpred, i is the predicted moisture ratio, Z is the number of parameters, and N is the number of observations.

Table 2 Mathematical models adopted in this research.

Model name	Model	Reference
Lewis	MR=exp(-kt)	[3]
Page	MR=exp(-kt^n)	[16]
Modified Page	MR= exp (-(kt)^n	[13]
Henderson & Pabis	MR=aexp(-kt)	[13]
Ademiluyi Model	MR=aexp(-(kt)^n)	[12]
Proposed Model	MR= 1/(a+kt^n)	Izionworu and Ojong

where k and a represent the drying constant and n are experimental constants.

2.3 Effective moisture diffusivity

The effective diffusivity was calculated from Fick's second equation of diffusion, considering an infinite slab geometry, constant moisture diffusivity and unchanging initial moisture spread:

$$MR = \frac{8}{\pi}{}_2\sum_{i=1}^{\infty}\frac{1}{(2n+1)}2\exp(\frac{-(2n+1)^2\pi^2}{4L^2}D_{eff}*t) \quad (5)$$

where L is the thickness of layer (m) and D_{eff} is the effective diffusivity (m^2/s). Equation (5) can be simplified by taking the first term, which gives:

$$MR = \frac{8}{\pi^2}\exp(-\frac{\pi^2 Deff*t}{4L^2}) \quad (6)$$

Equation (6) is numerically evaluated for Fourier number, $F_0 = D_{eff} \cdot t/4L^2$, for diffusion, revised as Equation (8) [17, 18]:

$$MR = \frac{8}{\pi^2}\exp(-\pi^2 F0) \quad (7)$$

Thus:

$$F0 = -0.101In(MR)-0.0213 \quad (8)$$

The effective moisture diffusivity was derived from Equation (8) as:

$$Deff = \frac{F_0}{\left(\frac{t}{4L^2}\right)} \quad (9)$$

3. Results and discussion

3.1 Moisture ratio of wire croton drying in the open air

The results of the experiment were used in the development of plots to show graphically the variation in drying over experimental time and functional drying parameters considering the environmental condition captured in the ambient temperature and humidity, as presented in Table 1.

The results of the drying curve fitted with CurveEpert Basic 22.3 presented in Figures 1 and 2 show the variation of MR of the Wire Croton sample with time over a period of 12 and 24 hrs' period, respectively. In the figures, it can be seen that the MR of the sample was reduced over time, although there was slight variation due to the influence of ambient temperature and humidity. Since the MR of the WC decreases as drying time rises, it shows that the MR, which is a function of the moisture loss of the sample, has a direct relationship with time, indicated by a decrease in moisture content as time increased, leading to a decrease in the MR of the sample with time.

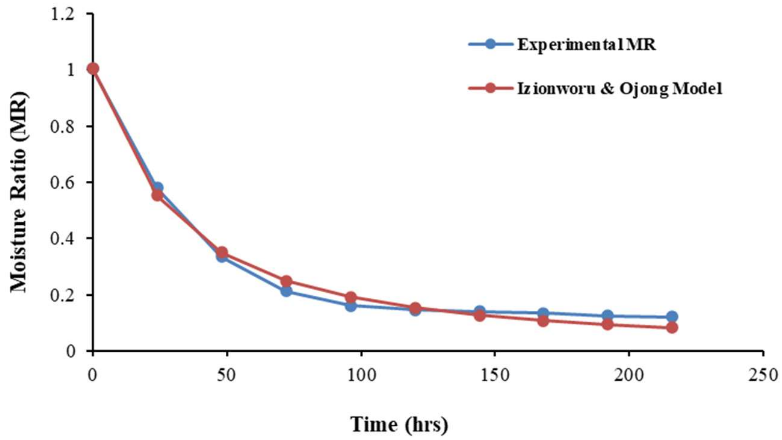

Figure 1 Moisture ratio against time for every 12 hrs' record of drying WC in open air.

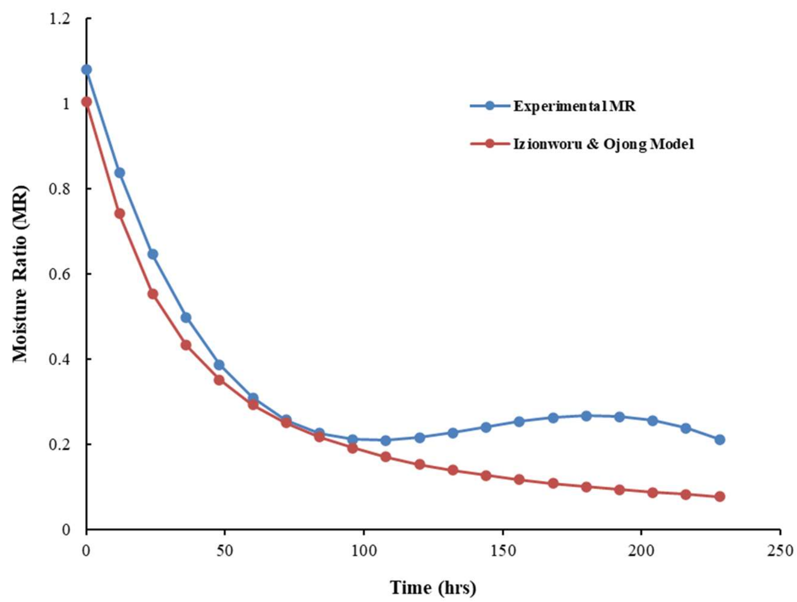

Figure 2 Plot of moisture ratio against time for every 24 hrs' record of drying WC in the open air.

3.2 Models comparison

Existing models, including Lewis, Page, Modified Page, Henderson and Pabis, and Ademiluyi, were compared for the best-fit model for the drying of WC. Figures 3 and 4 show the fitting results of these models, along with a newly proposed Izionworu and Ojong Model. Origin 08 software was used to determine regression parameters RMSE and select the best fitting model. The Izionworu & Ojong model had the lowest RMSE values of 0.104 and 0.029 for 12-hour and 24-hour intervals, respectively, and high R-squared values of 0.988 and 0.918. This model is considered the best for predicting the WC drying rate. The decrease in MR over time indicates the falling rate period, influenced by moisture movement and changing ambient conditions. Comparisons between experimental results and models showed that the proposed model had faster initial moisture loss. The Page model deviated by increasing the MR with drying time, while the Henderson and Pabis model had an initial MR different from 1. The proposed model better predicted the drying rate, with faster initial moisture loss and minimal losses over time. The Ademiluyi model also performed well with an R-squared value of 0.851 and an RMSE of 0.097.

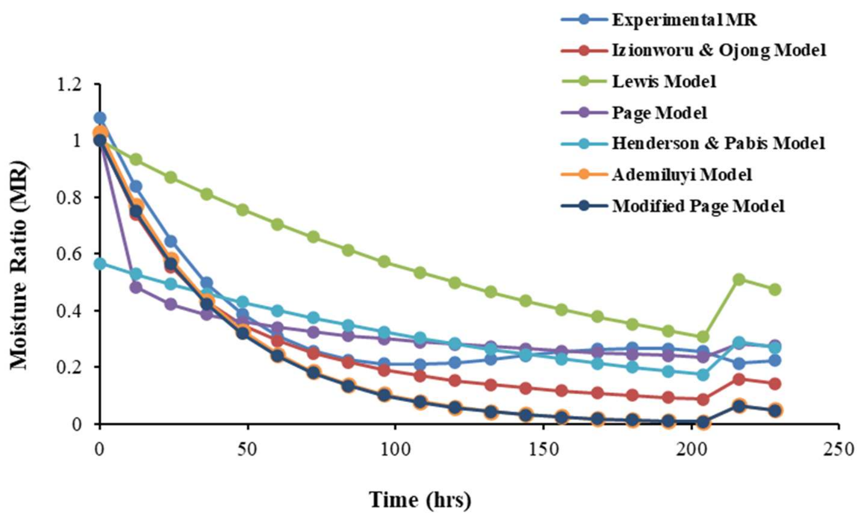

Figure 3 Fitting experimental and different predictive models for the moisture ratio of WC drying at 24 hrs intervals in the open air.

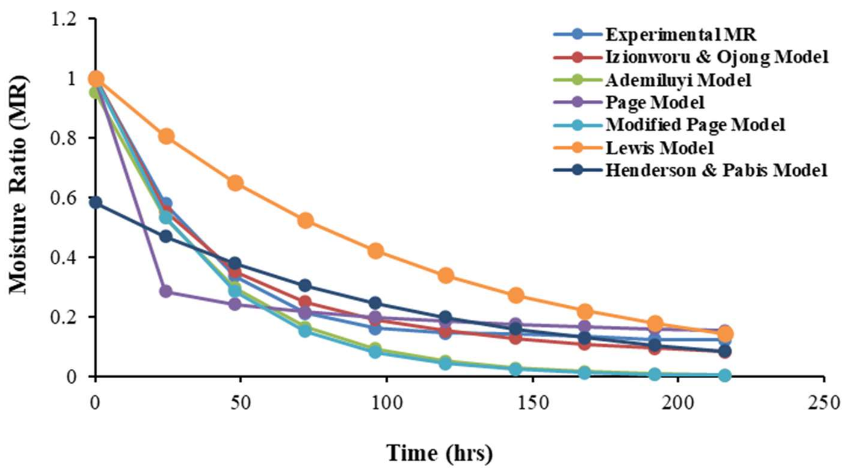

Figure 4 Fitting experimental and different predictive models of moisture ration of WC drying at 12 hrs intervals in the open air.

Table 3 Regression analysis of Wire Croton sample after every 12 hrs.

Time (hr)	Model name	Regression parameter				
		a	*K*	*n*	R^2	RMSE
	Lewis	-	0.0058	-	0.539	0.277
12	Page	-	0.40	0.2413	0.525	0.115
	Modified Page	-	0.0238	1	0.954	0.173
	Henderson & Pabis	0.567	0.0058	-	0.539	0.163
	Ademiluyi	1.028	0.024	1	0.954	0.170
	Izionworu & Ojong	0.995	0.018	1.193	0.988	0.104

Table 4 Regression analysis of Wire Croton sample after every 24 hrs.

Time (hr)	Model name	Regression parameter				
		a	*k*	*n*	R^2	RMSE
	Lewis	-	0.009	-	0.801	0.218
24	Page	-	0.697	0.185	0.448	0.113
	Modified Page	-	0.142	0.185	0.448	0.101
	Henderson & Pabis	0.583	0.009	-	0.801	0.162
	Ademiluyi	0.954	0.0244	1	0.851	0.097
	Izionworu & Ojong	1.635	0.035	1	0.918	0.029

3.3 Wire croton diffusion rate

The effectiveness of the diffusion rate of moisture from wire croton is dependent on time, which in turn depends on *MR*. Figure 5 shows that the effective diffusion rate decreases suddenly from 0 to -0.0018 when the time of diffusion increases from 0 hr to 20 hrs and then increases sharply to 0.0014 and then increases in a zigzag-like fashion to 0.002 and decreases to 0.0013 when the time further increases from 20 hrs to 23 hrs and then from 23 hrs to 72 hrs and further to 204 hrs. This implies that the diffusion rate is effective overall at a drying time of range 25 hrs to 72 hrs, which agrees with the proposed model developed for the drying of wire croton. In Figure 6, the effective diffusion rate increases suddenly from 0 to 0.038 when the time of diffusion increases from 0 hr to 25 hrs and then decreases exponentially to 0.023 and almost becomes steady to 0.020 when the time further increases from 25 hrs to 72 hrs and then to 216 hrs. This implies that the diffusion rate is effective at a lower drying time from 0 hr to 50 hrs, which agrees with the proposed model developed for the drying of wire croton. This behavior can be attributed to rapid moisture loss because of the initial higher quantity of moisture in the leaf. The humidity of the air region just above the leaves is indicative of the fluctuation in the diffusivity, suggesting that a regulated humidity in an open-air environment will improve the drying rate.

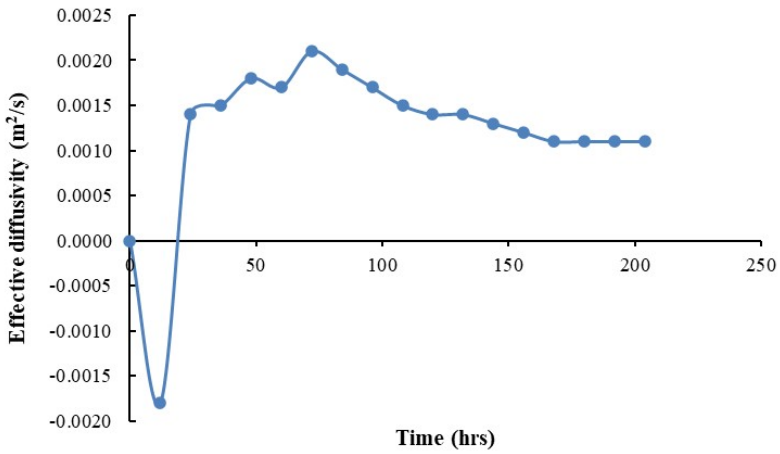

Figure 5 Effective rate of diffusion versus time for every 12 hrs' record of drying Wire Croton in open air.

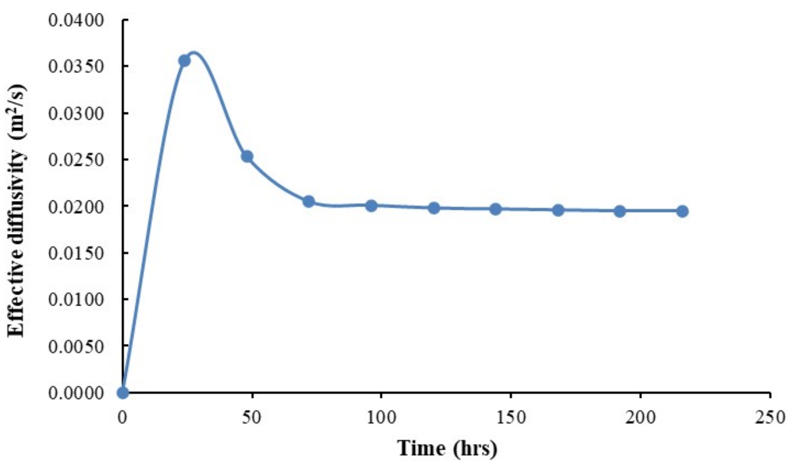

Figure 6 Effective rate of diffusion versus time for every 24 hrs' record of drying Wire Croton in open air.

The effective moisture diffusivity rate of 3.648×10^{-5} m^2/s and 2.432×10^{-5} m^2/s at 12 hrs and 24 hrs tests, respectively indicate an effective moisture diffusivity rate range that is higher than the effective moisture diffusivity rate for sawdust which was reported [24] as from 9.38×10^{-10} to 1.38×10^{-9} m^2/s under isothermal conditions. The effective diffusivity of olive leaves using microwave, vacuum, and oven drying under different steady temperature values varied from 10^{-9} to 10^{-12} [25], while the effective moisture diffusivity for mint leaves was observed to increase with the increase in drying air temperature and ranged from 1.2325×10^{-10} to $2.6568\times10^{-}$

^{10} m^2/s. The effective moisture diffusivity of garlic cloves under convective drying varied from 1.29 to 31.68 × 10^{-10} m^2/s [26], and that of onion slices varied from 0.21×10^{-10} to 1.57×10^{-10} m^2/s under a thin-layer radiation drying [27]. It is safe to conclude that the effective moisture diffusivity rate of biologically active dried products is at a range from 10^{-9} to 10^{-12} as specified [28]. Wire Croton size of 1x10 cm is quite small and enhanced the diffusivity. This agrees with the previous report that the size of leaves affects their drying time [29]. The high diffusivity also suggests a laminar airflow in the laboratory during the drying period, which agrees with the work of Babu et al. [19] and Arslan and Musa [30]. The humidity, as reported in Table 1, is favorable for high diffusivity, as reported by Babu et al. [19]. The temperature of the environment also affects the diffusivity as the laboratory is in a temperate region where the temperature range as reported in Table 1, was within 28-33°C; the results are in agreement with previous published reports by Bensebia and Allia Premi, Chraka et al. [31] and Premi et al. The temperature stagnated the laminar air, leading to increased diffusivity.

The method used by Chauhan et al. [32] was employed to validate the proposed Izionworu and Ojong two-term model for natural convective drying of WC. So, the calculated R^2 value of 0.988 and 0.918 for 24 hrs and 12 hrs, respectively from the use of Izionworu and Ojong's model were compared with other two-term models with the R^2 value of 0.896 and 0.826 calculated using Wang and Singh reported by Khanali et al. [33] and Midilli and Kucuk [34] correspondingly. The proposed Izionworu & Ojong model fits well. This is so because for both time intervals, the data fitted well as R^2 values resulting from the use of the proposed Izionworu & Ojong model is greater than the R^2 values calculated from the use of the Wang and Singh model and Midilli and Kucuk model [34]. Also, very low percentage deviation values 0, 7.1, 10.3, 2.5; and 19.6, 11.1, when validated with the existing models, proved that the proposed Izionworu and Ojong model is highly reliable and effective for both drying time intervals of WC.

In this study, the following limitations are identified – (1) low-temperature and convective heat transfer is considered to occur at the external surface of the porous medium and the drying air surrounding it. (2) Isothermal drying conditions are considered for the proposed model. (3) Relative moisture content is considered as the driving force for the process and (4) Regression kinetic model was thought of for the description of the kinetic model. The study assumed that at time zero, (i) the leaves are at uniform temperature and moisture content distributions, (ii) the side and bottom surfaces of the leaves had been insulted as it were, and dimensions in the other directions are sufficiently large that heat transfer and moisture diffusion may be considered as diffusion only through x axis, and (iii) the study also assumed that the porous bodies are continuous slab during the drying period and compressibility effects are negligible.

Drying at ambient temperatures has lower effective moisture diffusivity, as reported in previous studies [34 - 36]. However, D_{eff} of Wire Croton is very high and this explains the good drying result noticed at very low temperatures. The low D_{eff} enables the retention of compounds such as flavonoid, phytate, tannin, phenol, alkaloids useful for corrosion inhibition and present in WC, as heat is a major factor for making them inactive [34]. In terms of error propagation, the RMSE values obtained from the proposed Izionworu and Ojong model at the 24 hrs and 12 hrs drying time intervals are compared with literature models (two-term models of Wang and Singh [33] and Midilli [35], with values of 0.0297, 0.0866, and 0.0711 respectively. The percentage deviations computed were 2.4, 250, and 66.5, and 18.5, 59.2, and 46.3, respectively for 24 hrs and 12 hrs drying time, indicating that very high deviations were noticed for the two-term models except the proposed model (with negligible deviations). Izionworu and Ojong model gave a 2.4% deviation, very negligible compared with two-term models (250% and 66.5%) for 24 hrs and 18.5% negligible deviation compared with two-term models (59.2% and 46.3%) for 12 hrs drying time intervals of Wang and Singh [33] and Midilli et al. [35] models respectively. The high errors (deviations) obtained for 24-hour data compared to 12 hrs drying time data imply that the drying of the WC at 12 hrs time intervals is best as it minimizes errors and is good for the process and the preservation of the WC [36].

4. Conclusion

The drying kinetic study and modelling of Wire Croton were carried out in this work using experimental and mathematical modelling methods at ambient temperature and humidity. The experimental results obtained were fitted to Lewis, Page, Modified Page, Handerson-Pabis and Ademiliyu's thin-layer models with regression coefficients of 0.539, 0.525, 0.954, 0.539 and 0.954, respectively, in the 12 hrs test reported and R^2 value of 0.801, 0.448, 0.448, 0.801 and 0.851, respectively for the 24-hourly test computation.

The experimental drying of WC under a shade indicates that WC can be dried under a shade in ambient conditions in the Niger Delta region of Nigeria. This saves the cost of using additional heat from any heating source. It also implies that WC can be safely dried in a shade, preventing contamination by dirt etc if dried under the sun in the open air. The overall implication is that the resulting biomass from WC dried in a shade results in extracts of WC to be used as inhibitors for metal corrosion control that retain its bioactive components yet are free of contaminants. Izionworu & Ojong proposed a polynomial model of degree 4 for this work, which was tested and the result was compared with existing models. The RMSE from Izionworu & Ojong's proposed model shows that this model has the least RMSE value of 0.104 and 0.029 at 12- and 24-hour measurement respectively, and R^2 results of 0.988 and 0.918 for 12- and 24-hour interval computation which is higher than all existing models considered in this study. Therefore, Izionworu & Ojong's proposed model is most suitable for the description of the drying behavior of wire croton over a 12- and 24-hour period in the tropical sub-Sahara environment.

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