MODELLING THE EFFECT OF TEMPERATURE ON DRYING MECHANISM OF CATFISH CRACKER

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ABSTRACT

The drying characteristics of 2 mm slices of formulated catfish cracker were investigated at 40, 50 and 60 °C and at a fixed air velocity of 1.5 m/s using a convective oven dryer. The samples were dried up to equilibrium moisture content. It was apparent from the results that the drying time reduced as the drying temperature increased. The experimental drying data of the formulated fish cracker were fitted to three semi-empirical thin layer drying models: Page's, Henderson and Pabis and Logarithmic. Statistical parameters of R^2, X^2 *and* $RMSE$ *were used to determine the suitability and for identification of best performing model for describing the drying kinetics of formulated catfish cracker slices. The Logarithmic model gave the best prediction when compared with other tested models. From the results obtained, two different falling rates periods were observed, and the effective moisture diffusivity (*$Deff$*) values were found to range between 2.26495 E-08 - 4.49344E-08 m*^{2}*/s and 4.21387E-09 - 8.7924 E-09 m*^{2}*/s for the first and second falling rate periods respectively. Increasing drying temperature also caused an increase in* $Deff$ *values. The activation energies were also found as 29.63 kJ/mole and 31.82 kJ/mole for first and second falling rate periods respectively. The Logarithmic model can, therefore, be employed to understand the mechanism of catfish cracker drying, the design of the drying process, and the design of drying equipment, prediction and control of the process.*

***Keyword**: Catfish, Cracker, Diffusivity, Activation Energy, Thin-layer modelling*

1. INTRODUCTION:

Fish is a high-protein, low-calorie food and an important source of omega-3 fatty acids and minerals, such as calcium and phosphorus (Okereke and Onunkwo, 2014). Fish is the most easily accessible animal protein especially in the developing countries. Nowadays, fish cracker, a product of fish utilization process, is gaining a widespread acceptability in Nigeria as an entertaining ready-to-eat snack. The snack is easy to make because it is sold as an intermediate food product leaving only the deep frying aspect to the consumer to perform. Fish cracker, also represents a route to which stunted fishes with low economic values have an economic benefit to the fish farmer (aqua culturist). Although fish cracker can be made in different ways, they are usually produced by mixing flesh of a specific fish, food starch, monosodium glutamate (MSG) and other spices. Other ingredients can be added to enhance better acceptability and improved properties; this is dependent on the producer (Mbaeyi-Nwaoha and Itoye, 2016). The ingredients after mixing are steamed/boiled, refrigerated, sliced into small piece and then dried first before frying (Mbaeyi-Nwaoha and Itoye, 2016; Nurul *et al.,* 2009). Drying of the cracker is necessary to reduce its moisture content for preservation, storage, marketing and post processing (deep frying). Most commercial and locally formulated (homemade) crackers conform to 1-3 mm slices (Mbaeyi-Nwaoha and Itoye, 2016; Netto *et al.,* 2014) perhaps to enhance fast drying of the product. In Nigeria, because of the availability of sun energy, drying of formulated fish cracker is usually done through sun drying which is not too predictable and constitutes a limitation in improving the fish cracker production from small scale to large scale production. The importance of the choice of drying route and optimization is, therefore, significant in fish cracker processing and development. In this sense, the understanding of products' drying kinetics, mathematical modelling, and evaluation of related thermodynamic parameters will improve the product quality and assists in equipment design.

Drying is a dual process that involves the internal penetration of the sample by heat energy and outward diffusion of moisture from the sample. Control of drying process in food or agricultural product is necessary to avoid unwanted product and destruction of micro nutrients. Drying is an energy intensive unit operation and long drying periods tend to increase the energy requirements for the production of the dry product (Agarry *et al.,* 2013). Hence process optimization is necessary for the economic drying of specific products. Drying in Africa is usually carried out through natural sun drying. However, because of its unhygienic implications, the use of aided technologies is rapidly becoming acceptable, especially with emerging strict laws on food processing from the government agencies. Drying of materials is a complicated process involving simultaneous heat and mass transfer (Okereke and Onunkwo, 2014). Generally, the drying process takes place in two stages; the first stage happens at the surface of the drying material at a constant drying rate and is similar to the vaporization of water into the ambient. The second stage drying process takes place with decreasing drying rate (Okereke and Onunkwo, 2014) as heat energy passes from the surface to the core of food.

Mathematical modelling and simulation of drying curves under different conditions are important to obtain a better control of drying unit operation and an overall improvement of the quality of the final product (Hazbavi and Samadi, 2013). The mathematical modelling allows the food researchers to choose the most suitable operating conditions either to describe the drying equipment or minimize the drying times for the final product specifications (Gamll, 2011). The thin-layer drying models can be categorised as theoretical, semi-theoretical and empirical models. The semi-theoretical model based on the theory and the drying kinetics experimental, is derived from the simplification of Fick's second law of diffusion or modification of the simplified model, which has been widely used to describe the drying characteristics (Guan *et al.,* 2013). Drying should also progress in a regular manner because spontaneous drying may lead to case-hardening of the sample. Page model had been investigated for studying the drying characteristics of some fruit and vegetables such as pepper (Akpinar *et al.*, 2003), apricot (Mirzaee *et al.,* 2010), purslane (Demirhan and Özbek, 2010) and mango fruits (Kabiru *et al.*, 2013). Other empirical models used for fitting kinetic data of agricultural products include Midilli model (Mirzaee *et al*., 2010), Wand and Singh model (Hamdami *et al.*, 2006; Mirzaee *et al.,* 2010), Logarithm, Henderson and Pabis, Tow term and Newton models (Mirzaee *et al.,* 2010). All the empirical models consist of drying rate and equation constants which must be obtained from the fitting of the experimental kinetic data to the models. The statistical parameters such as coefficient of determination (R^2), chi-square (χ^2) and root mean error (RMSE) are frequently used for determining the suitability of each model in describing the experimental kinetic data.

The moisture diffusivity is an important parameter in material characterization, and the knowledge of moisture diffusivity enables the proper choice of drying and or process conditions for specific foods. Molecular diffusion is the main water transport mechanism in dehydration and to predict the water transfer in food materials, diffusion models based on Fick's second law are frequently used (Okereke and Onunkwo, 2014). Basically, in the application of Fick's law to evaluate moisture diffusivity of infinite slab of thin layer, assumptions such as moisture migration being by diffusion, one-dimensional moisture movement, uniform initial moisture distribution, negligible shrinkage, constant moisture diffusivity, and negligible external resistance to heat and mass transfer are usually made. Effective moisture diffusivity describes all possible mechanisms of moisture movement within the foods, such as liquid diffusion, vapour diffusion, surface diffusion, capillary flow and hydrodynamic flow (Okereke and Onunkwo, 2014). It is therefore expected that moisture diffusion will increase with moisture content especially when the sample is fresh, progressively decrease and later remain constant. Other frequently studied products' thermal property frequently measured is activation energy and this is because of its usefulness in ideal dryer design (Aghbashlo *et al.*, 2008).

The objectives of this study are as follows. For catfish cracker dried in a conventional oven, (1) Obtaining the drying kinetic data for mass/moisture transfer during the convective oven drying process of catfish cracker, (2) determination of the thin layer drying model that best fits the drying data, (3) estimation of the effective diffusivities at drying temperatures and (4) estimation of the activation energy of the drying operation. The study aims to understand the drying behaviour of catfish cracker for process design, equipment design, and optimization.

2.0 MATERIALS AND METHODS

2.1 Materials

Fresh catfish of about 500 g was purchased from a local food market in Lagos, Nigeria. Other ingredients such as corn starch, pepper, salt and magi were purchased from a supermarket in Lagos. The dryer used for the experiment is a convective oven dryer equipped with fan, temperature regulator and timer. The dryer is 1m by 2 m rectangular shaped drying chamber with relatively high capacity loading. The dryer was also equipped with two strategically positioned heating elements (1.5 kW) at the top and bottom for total heat coverage of the chamber. The door of the dryer was made of transparent material for easy visual monitoring of the product. A primerie model weighing balance with accuracy of 0.01 g was used for the experiment. Knife was used for fish deboning, degutting and filleting. Pestle and mortal was used for mashing of the deboned fish flesh. Steaming pan and refrigerator were also used for the production process.

2.2 Methods

2.2.1 Preparation of Catfish Cracker Sample

The recipe and method of Okereke and Onunkwo (2014) were adopted for the preparation of catfish cracker. The pre-fish processing was done according to the method of Okereke and Onunkwo (2014). In brief, the catfish was washed, gutted, filleted and de-headed manually using a sharp knife. The flesh was further washed with running tap water. 35 g of filleted and deboned catfish flesh was mashed using the attrition pepper grinder and was mixed with 25 g of corn starch, a cube of magi, 0.01 g of pepper and 0.05 g of salt. The mixture was rolled out into a cylindrical shape using a tin mould and then steamed for 45 minutes. After, samples were kept in the refrigerator at -4 ^{O}C before the drying experiments. Multiple 5 g of thinly sliced fish cracker (2 mm) samples were made and selected for each batch of the experiment. The experiment was designed to be a one at a time experiment through which variable inputs temperature (40, 50 and 60 ^{O}C), air velocity of 1.5 m/s and constant thickness of 2 mm (based on local producers' standard practices) were used for the investigation. Moisture loss output/responses were taken at constant intervals in agreement with previous studies.

2.2.2 Determination of Initial Moisture Content

The initial moisture content of the fish cracker was determined by using oven drying method (AOAC, 1984). About 5 g of samples were oven dried at 70 ^{O}C for 12 h. The initial moisture content was established on a dry basis (d.b.). The experiments were conducted in triplicates and the initial moisture content of the fish cracker was reported as the average of three experimental trials. The relationship below (Equation 1) was used to establish the initial moisture content.

$$Initial\ Moisture\ Content\ (db.) = \frac{Wet\ Weight - Dried\ Weight}{Dried\ Weight} X\ 100\ (\%) \quad (1)$$

2.2.3 Determination of Moisture Loss and Associated Parameter

The samples were defrosted by allowing the fish cracker slices to reach the room temperature (25^{o}C) before drying experiments proceeded. Hazbavi and Samadi (2013) method was used for the drying experiment, where the change in moisture was recorded at an interval of time. The effect of temperature variation was studied as a function of time at an air velocity of 1.5 m/s. The selected temperatures were 40, 50 and 60 ^{O}C. However, before the drying experiment, the dryer was allowed to work at the set experimental temperature for 30 min such that the temperature throughout the oven chamber was uniform. At each set temperature, the 5 g of 2 mm thinly sliced sample is placed on a mesh holder and inserted in the oven chamber. The moisture loss was recorded at 5 min time intervals during the experiment with an accuracy of 0.01 g. The investigation continues until there was no difference between four successive recordings (equilibrium). The drying process was carried out to final moisture content of about 2 - 2.3 % from an initial moisture content of about 151.5 % (db.).

Furthermore, the moisture loss values were converted to a suitable format for use in kinetics modelling. Specifically, instantaneous moisture losses were converted to moisture ratio using the established relationship and assumptions. The relationship is as in Equation 2

$$MR = \frac{Mt - Me}{Mo - Me} \quad (2)$$

from the relation, MR represent moisture ratio, M_t represents the moisture content of the product after drying time t, M_e represents the stable or equilibrium moisture content of the product and M_0 represents the initial moisture content of the sample. It can be easily seen that the equation will naturally reduce to the ratio of present moisture content to original moisture content as in Equation 3.

$$MR = \frac{Mt}{Mo} \quad (3)$$

Drying rate was also determined to understand the rate at which the material losses moisture over time. The mathematical relationship used for calculating the drying rate is expressed in Equation (4).

$$DR = \frac{Xt + \Delta t + Xt}{\Delta t} \quad (4)$$

Where$Xt + \Delta t$, is the moisture content at a future time, Xt is the moisture content at the present time and Δt is the change in time.

2.2.4 Fitting of Empirical Models

Three semi-empirical models were fitted to the experimental moisture ratio data to better understand the mechanism of drying of the developed catfish cracker. The used models were Page model (Equation 5), Henderson and Pabis model (Equation 6) and Logarithmic model (Equation 7).

$$MR = Exp.(-K.t^n) \quad (5)$$

$$MR = a.Exp.(-K.t) \quad (6)$$

$$MR = b.Exp(-K.t) + C \quad (7)$$

where n and b are exponent specific to each model: a, K and C are model constants and t is the drying time

Curve fittings for the chosen semi empirical models were performed using the solver function in Microsoft Excel adopting the generalized reduced gradient (GRG2) nonlinear optimization code to determine the drying parameters. The suitability of each model was judged using the statistical parameter values of coefficient of determination (R^2) (Equation 8), root mean square error (RMSE) (Equation 9) and reduced chi square (X^2) (Equation 10). A high value of coefficient of determination coupled with low values of root mean square and chi-square signifies a better model.

$$R^2 = 1 - \left(\sum_{i=1}^{N} \frac{(\text{MRpre,i} - \text{MRexp,i})^2}{(\text{MRpre,i} - \text{AverageMRexp})^2}\right) \quad (8)$$

$$X^2 = \frac{\sum_{i=1}^{n}(\text{MRexp,i} - \text{MRpre,i})^2}{N-Z} \quad (9)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(\text{MRexp,i} - \text{MRpre,i})^2}{N}} \quad (10)$$

where, N is the number of observations, Z is the number of constants, MRexp and MRpre are the experimental and model predicted moisture ratios, respectively.

2.2.5 Determination of Coefficient of Diffusivity and Activation Energy

Fick's second law of diffusion was used to calculate the moisture diffusivity of catfish cracker. The analytical solution of second Fick's law given by Rayaguru and Routray (2011) is represented in Equation 11 as:

$$MR = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{2n+1} \exp\left[-(2n+1)^2 \frac{\pi^2}{4L^2} Deff * t\right] \quad (11)$$

where MR is the moisture ratio, $Deff$ is the effective moisture diffusivity in m^2s^{-1}; L is the half thickness of the product in meter (m) and t is time in second (s). For long drying period, the Equation (11) can be simplified to Equation (12) as follows:

$$ln\,(MR) = ln\,(\frac{8}{\pi^2}) - Deff\,(\frac{\pi}{2L})\,\hat{}2 * t \quad (12)$$

The effective moisture diffusivity was determined using Fick's law during the falling rate period. Briefly, the effective diffusivity was determined from the slope (Equation 13) of a straight line obtained by plotting natural logarithm of experimental drying data ($ln(MR)$) against time (t).

$$Slope = \frac{\pi^2\, Deff}{4L^2} \quad (13)$$

The relation between temperature and the effective moisture diffusivity can be described by an Arrhenius-type relationship in Equation 14 (Guan *et al*., 2013)

$$Deff = D_o\, Exp\,(\frac{E_a}{RT_a}) \quad (14)$$

where D_o is the pre-exponential factor for Arrhenius equation (m^2/s) and E_a is the activation energy for moisture diffusion (kJ/mole), R is the gas constant (kJ/mole.K) and T_a is the absolute temperature in kelvin (K). The activation energy was obtained from the slope (Equation 15) of the straight line by plotting $ln(Deff)$ versus the reciprocal of the temperature ($1/T_a$)

$$Slope = -\frac{Ea}{R} \quad (15)$$

3 RESULT AND DISCUSSION

3.1 Effect of Drying Temperature and Time on Catfish Cracker Moisture Ratio

Figure 1 shows the effect of drying temperature and time on the moisture ratio (MR) of the formulated catfish cracker at an air velocity of 1.5 m/s. As expected, the MR of the formulated catfish cracker decreased with increased drying time. This was although pronounced at the beginning of the experiment when moisture was being rapidly removed from the surface of freshly prepared cracker due to the availability free moisture which was rapidly removed and became less obvious as the drying experiment progressed. The decreased moisture removal at the latter end of the experiment may be as a result of decreased moisture content or case hardening of the catfish cracker which prevented the heat from entering the core of the product. The same observation was reported for the drying behaviour of some agricultural products such as onion slices (Revaskar *et al.*, 2014), kiwi fruit slices (Shahi *et al.*, 2014), *Pandanus amaryllifolius* leaves (Rayaguru and Routray, 2011), Okra (Afolabi and Agarry, 2014) and Fish fillets (Ikrang *et al.*, 2014).

Drying temperature also had a great effect on the drying behaviour of the cracker. The relative positions of the drying profiles showed that the MR decreased with increased temperature due to increased drying rate. This is in accordance with the report of Nag and Dash (2016) on mathematical modeling of thin layer drying kinetics and moisture diffusivity study of elephant apple. For instance, when the drying temperature was increased from 40 to 60 ^{O}C, the drying time reduced by 49 % which implied a faster moisture removal at the higher temperature. However, too high a temperature will lead to case hardening phenomenon, denaturing and even burning.

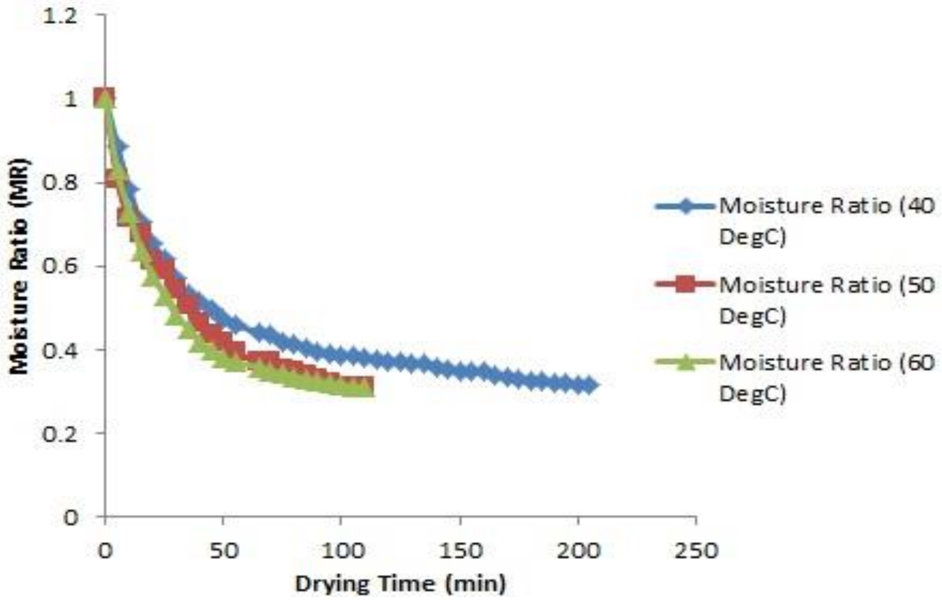


Figure 1: Graph of Moisture Ratio versus Drying Time for the drying of fish cracker

Figure 2 shows the graph of drying rate against drying time. The drying rate increased rapidly at the onset of drying and reached the peak in 5 min before it later fell. This indicated that 5 min was enough to remove all the free moisture on the surface of catfish cracker before the further inward-to-outer moisture removal began. The drying rate however, continued to reduce afterwards and this may be due to the slow internal outward moisture movement which implied that catfish cracker may not be too porous for easy movement of moisture. The drying rate clearly showed its dependency on temperature and it increased as the temperature increased.

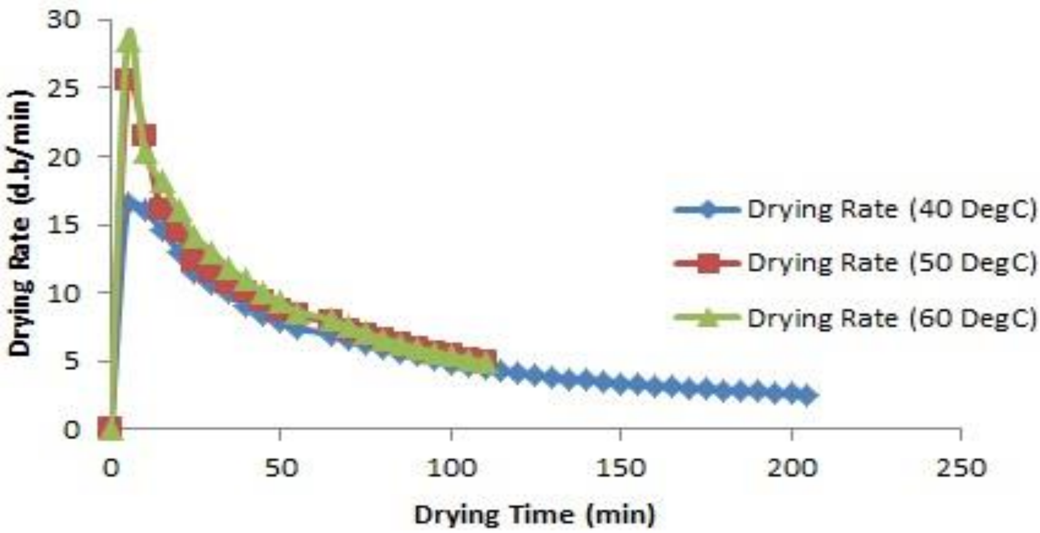


Figure 2: Graph of Drying Rate versus Drying Time for the drying of fish cracker

3.2 Modelling of the Drying Kinetics

In order to describe the effect of temperature on the kinetics of the formulated fish cracker, three semi-empirical thin-layer drying kinetic models were considered as detailed in Equations 5 – 7. Among the examined models, Logarithm model was judged the most appropriate for the description of experimental kinetic data at all temperatures with highest R^2 values, and lowest χ^2 and RMSE values. The estimated model parameters at different drying conditions are as illustrated in Table 1.

Table 1: Comparison of different models with parameters for drying of formulated catfish cracker

Model Name	Temperature (^{O}C)	Constants	R^2	χ^2	$RMSE$
Page	40	**k**= 0.1825 **n** = 0.528	0.9413	0.00213	0.04503
Henderson and Pabis	40	**a** = 0.7661 **k** = 0.029	0.7901	0.00547	0.07213
Logarithmic	40	**b** = 0.7352 **k** = 0.1578 **c** = 0.3412	0.9909	0.00024	0.01501
Page	50	**k** = 0.1437 **n** = 0.722	0.9599	0.00146	0.03646
Henderson and Pabis	50	**a** = 0.8951 **k** = 0.061	0.9202	0.00291	0.05148
Logarithmic	50	**b** = 0.7775 **k** = 0.1744 **c** = 0.2973	0.9903	0.00037	0.01797
Page	60	**k** = 0.1672 n = 0.684	0.9265	0.00271	0.04970
Henderson and Pabis	60	**a** = 0.8766 **k** = 0.064	0.8662	0.00500	0.06745
Logarithmic	60	**b** =0.8454 **k** = 0.233 **c** = 0.3137	0.9986	0.00005	0.00685

Observations from Table 1 indicate that the parameters of Logarithm model increased as temperature increased. The implication is that with the increase in temperature, the drying curve becomes steeper meaning an increase in drying rate. From the graphs in Figure 3, it is qualitatively obvious that the Logarithmic model outperformed the other models that were used in the modelling of the experimental data.

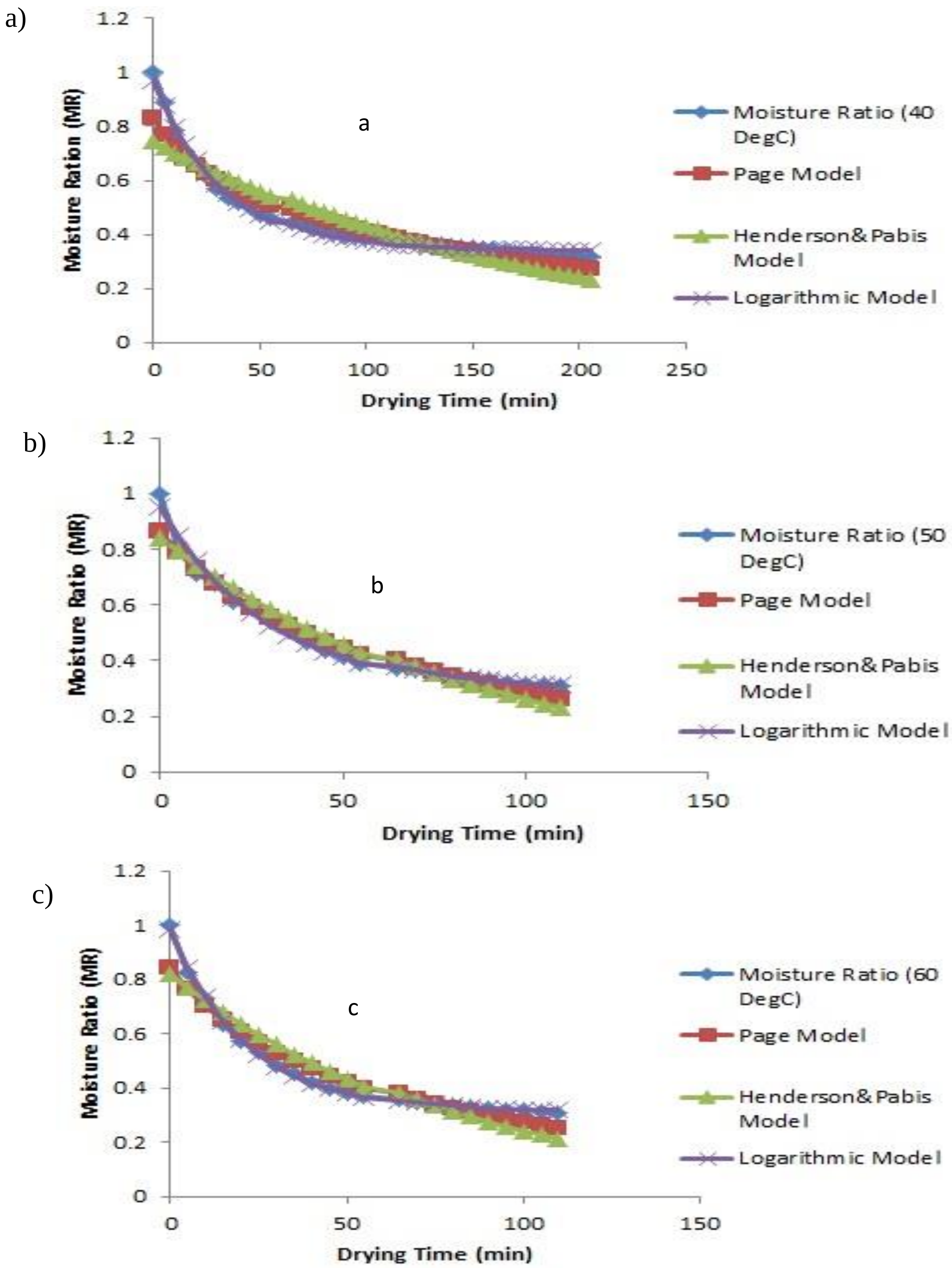


Figure 3: Prediction of experimental moisture ratio as a function of drying time using thin layer semi empirical drying models at (a) 40 ^{O}C (b) 50 ^{O}C (c) 60 ^{O}C.

3.3 Effect of Temperature on the Coefficient of Moisture Diffusion

The diffusivity was evaluated through linear regression from the slope of the ln (MR) versus time relationship as depicted in Figure 4. From Figure 4, it is apparent that the drying of fish cracker at all the three temperatures occurred in two distinct falling rate periods. A similar observation was reported by Motevali *et al.* (2012) in the thin-layer modelling of Jujube. Therefore, for a specified constant temperature, two different diffusivities must be established for the two observed falling rates periods accordingly.

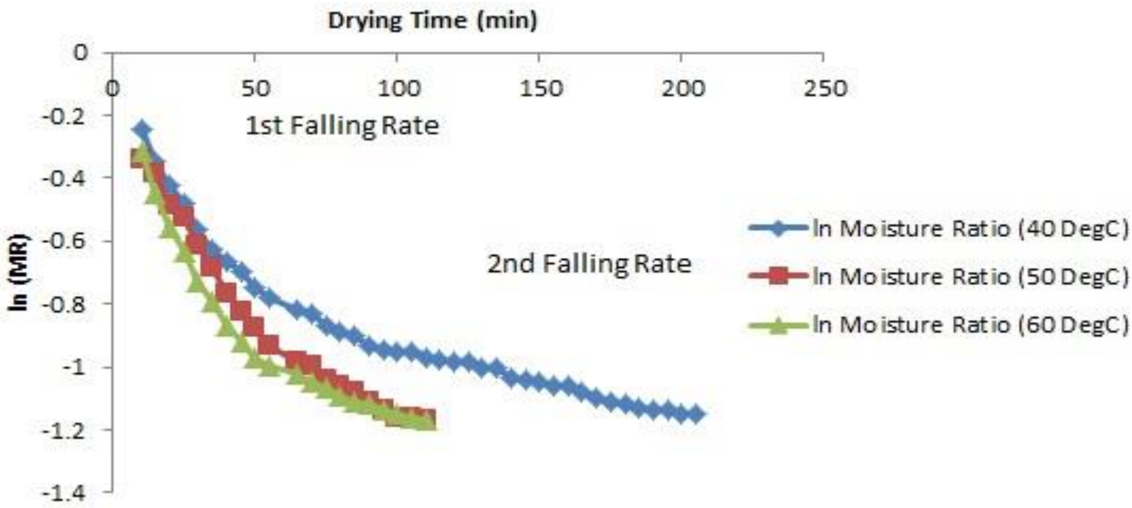


Figure 4: Plot of ln (MR) against Time

The effective moisture content values for various drying temperatures were determined and presented in Table 2. The range of moisture diffusivity values for the first falling rate period was from 2.26495E-08 to 4.49344E-08 m^2/s and that for the second falling rate period varied from 4.21387E-09 to 8.7924E-09 m^2/s. Observations indicated that the moisture diffusivity of the first and second falling rate periods increased with increased temperature. A similar range of moisture diffusivity was reported for agricultural products. Mariem *et al.* (2014); reported a range 10^{-9} and 10^{-8} m^2/s for tomato samples, while Guan *et al*. (2003) reported a range of 10^{-8} - 10^{-12} m^2/s for food materials.

Table 2: Diffusivity values at different temperatures and rate periods

	Diffusivity (m^2/s)	
Temperature (^{O}C)	**1st Falling Rate Period**	**2nd Falling Rate Period**
40	2.26495E-08	4.21387E-09
50	3.051E-08	5.8751E-09
60	4.49344E-08	8.7924E-09

Equations 16 and 17 are the regressed relationships between effective moisture diffusivity and drying temperatures for the first and second falling rates respectively.

$Deff$ = 3E-11T^2 - 2E-09T + 6E-08 first falling rate (16)

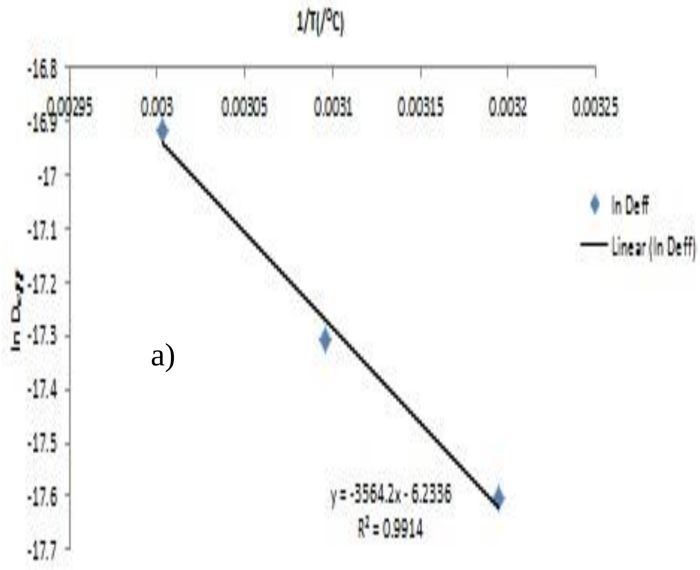
$R^2 = 1$

$Deff$ = 6E-12T^2 - 4E-10T + 1E-08 Second falling rate (17)

$R^2 = 1$

3.4 Effect of Temperature on the Activation Energy

The activation energy (Ea) was determined from the slope of the linearized Arrhenius equation. A graph of ln ($Deff$) versus 1/ T was made and Ea was evaluated according to the method of Mariem and Mabrouk (2014). Figures 5 showed the relationship between ln($Deff$) versus 1/ T for the first and second falling rate periods respectively.



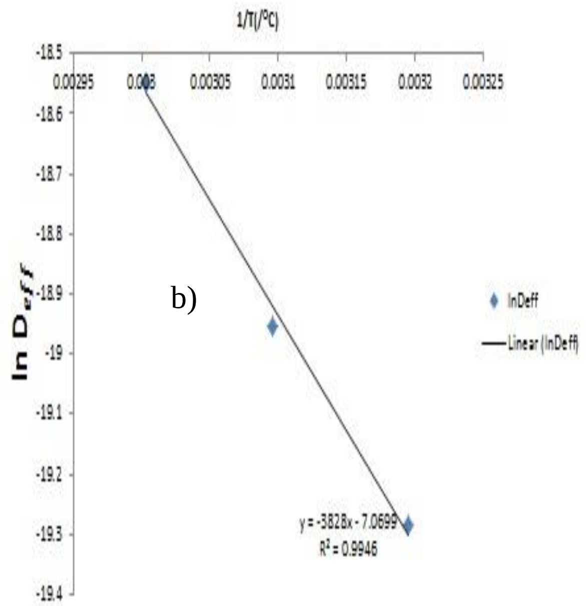


Figure 5:Graph of ln ($Deff$) versus inverse of absolute temperature for (a) first falling rate period (b) second falling rate period

Ea for the first and second falling rate periods as previously shown in Figure 4 was determined to be 29.63 kJ/mol and 31.82 kJ/mol respectively. The results were in close agreement with the values of 24.94 kJ/mol and 28.40 kJ/mole reported for tomato slices and green peas respectively (Mariem *et al.*, 2014).

4 CONCLUSION

The thin layer drying of formulated fish crackers was investigated. The experimental drying data were fitted to some semi-empirical thin layer models. Thermodynamic parameters such effective moisture diffusivity and activation energy were also determined. The moisture ratio of the formulated catfish cracker decreased with increased drying time and temperature. Drying proceeded in falling rate periods only till equilibrium was reached. The Logarithmic equation had the highest R^2 and lowest chi-square and RMSE values and was the most suitable thin layer model for describing the drying kinetics of the formulated catfish cracker. The effective moisture diffusivities and the activation energies were found to be in the range 2.26495E-08 - 8.7924E-09 m^2/s and 29.63 - 31.82 kJ/mol for the first and second falling rate periods respectively. The results obtained can be well used for analysis, design, understanding, and prediction of mechanisms involved in the drying of catfish cracker.

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Nomenclature

a, b, c	Model coefficient
d.b.	Dry basis
SE	Standard error
RMSE	Root mean square error
SST	Sum of square
k	Drying rate constant (min^{-1})
M	Moisture content (% d.b.)
MR	Moisture ratio
M_0	Initial moisture content (% d.b.)
M_e	Equilibrium moisture content (% d.b.)
$MR_{exp,i}$	ith Experimental moisture ratio
$MR_{pre,i}$	ith Predicted moisture ratio
n	Exponent
R^2	Coefficient of determination
t	Time (min)
D_{eff}	Effective diffusivity, m^2/s
D_0	Pre-exponential factor of Arrhenius equation, m^2/s
E_a	Activation energy, kJ/mol
k_0	Slope
R	Gas constant
L	Half slab thickness

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