

Effects of Drying Temperature on The Effective Coefficient of Moisture Diffusivity and Activation Energy in Ibadan-Local Tomato Variety (*lycopersicum esculentum*)

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Abstract

A study of the mechanism of mass transfer phenomena of Ibadan-local variety was carried out. Ibadan-Local tomato varieties pre-treated in a binary (sugar and salt) osmotic solution of concentration (45/15^{o}Brix), solution temperature (30, 40, 50^{o}C), was studied by developing a mathematical model to describe the Water Loss (WL) and Solid Gains (SG). Drying was monitored at three temperatures (40, 45 and 50^{o}C) until equilibrium weight was achieved using the oven-dry method. Five thin layer drying models (Exponential, Henderson & Pabis, Page, Modified Page and Logarithmic) were compared and fitted into the experimental values of the non-linear moisture ratio; MR. The diffusion coefficient and activation energy were determined using the Arrhenius equation. Drying occurred in the falling rate phase and different models fit at different temperatures. Calculated values of effective moisture diffusivity varied from 1.17-3.51x10^{-8} to 1.25-3.13x10^{-8} and activation energy varied from a maximum of 52.61KJ/mol in treated to 46.81 KJ/mol in untreated tomato. At all temperatures, effective coefficient of moisture diffusivity and activation energy values was higher in osmosized tomato

Keywords: *Osmotic dehydration, Water loss, Solid gain, Effective moisture diffusivity and Activation energy.*

1.0 INTRODUCTION

Fruits and vegetables contain more than 75% of water and tend to deteriorate quickly if not properly stored (FAO, 2007). Generally, the fruits cannot be stored for a long period without deterioration unless in dry form, dehydration reduces moisture in food to a level that inhibits the microbial growth that causes deterioration.

Drying is the most common form of food preservation and it extends the shelf-life of food (Raji *et al*. 2010). The major objective in drying agricultural product is the reduction of the moisture content to a level, which allows safe storage over an extended period, it brings about substantial reduction in weight and volume, minimizing packaging, storage and transportation costs (Okos *et al*. 1992). Traditionally, tomatoes are sun-dried and this usually takes time depending on the variety of tomato, the humidity in the air during the drying process, the thickness of the slices or pieces, and the efficiency of the dehydrator or oven (Kaur *et. al*. 1999). Sun drying is a common method that is naturally simple and requires less capital. However, it is time consuming, prone to contamination with dust, soil, sand particles birds and insects and it is weather dependent. Other drying methods that could be explored include solar and the oven method.

These drying methods (i.e. solar and oven-drying) have however proved from different studies to be deficient hence, the introduction of a dehydration method called Osmotic Dehydration which is capable of reducing the moisture content of foods by 50%. Fruits and vegetables are subjected to pre-treatment before drying them with a view to improving their drying characteristics and minimizing adverse changes during drying. Such pre-treatment may include alkaline dips, sulphiting, osmotic dehydration, etc. However, pre-treatment excluding the use of chemicals may have greater potential in food processing (Ade –Omowaye *et al*. 2003). This explains why osmotic dehydration is used as a pre-treatment/pre-processing method to be followed by other drying methods.

The use of conventional tray dryers or vacuum dryers for fruits produce are wholesome, nutritious and palatable products in its own right but has not in general found popular acceptance because the final product does not have the flavour, colour and texture of the original fruit even after re-hydration (Bongirwar and Screenivasan 1977).

Drying of tomatoes for many years back has been through sun drying. Sun dried tomatoes are however known to have practically all the organoleptic properties removed and its success is a function of the intensity of sunlight that is made available, hence there is the need to carry out research work on a good preservation method that will meet consumer's taste.

Drying kinetics is greatly affected by their velocity, air temperature, material thickness and others (Ereturk and Ereturk, 2007). Some researcher have studies the moisture diffusion and activation energy in the thin layer drying of various agricultural products such as Seedless grapes Plums , grapes, candle nuts , potato slices and onion slices. Although much information has been given on the effective moisture diffusivity and activation energy for various agricultural products, no published literature is available on the effective moisture diffusivity and activation energy data for Ibadan-Local tomato during drying. The knowledge of effective moisture diffusivity and activation energy is necessary for designing and modeling mass transfer processes such as dehydration or moisture absorption during storage.

2.0 Theoretical Consideration

Determination of Effective diffusivity coefficients

Drying process of food materials generally occurs in the falling rate period (Wang & Brennan, 1992). Determining coefficient used in drying models is essential to predict the drying behaviour. Mathematical modeling and simulation of drying curves under different conditions is important to obtain a better control of this unit operation and overall improvement of the quality of the final product. To predict the moisture transfer during the falling rate period, several mathematical models have been proposed using Fick's second law. Application of Fick's second law is usually used with the following assumptions (Crank, 1975).

(i) Moisture is initially distributed uniformly throughout the mass of a sample.

(ii) Mass transfer is symmetric with respect to the center

(iii) Surface moisture control of the sample instantaneously reaches equilibrium with the condition of surrounding air

(iv) Resistance to the mass transfer at the surface in negligible compared to internal resistance of the sample

(v) Mass transfer is by diffusion only and

(vi) Diffusion coefficient is constant and shrinkage is negligible.

$$MR = \frac{M}{M_0} = \frac{8}{\pi^2} \sum_{n-1}^{\infty} \exp\left[-\frac{(2n-1)^2 \pi^2}{4L^2} Dt \right] \quad (1)$$

Where MR is moisture ratio, M is the moisture content at any time (kg water / kg dry matter), M_0 is the initial moisture (kg water / kg dry solid), n = 1, 2, 3 …… the number of terms taken into consideration, t is the time of drying in second, O is effective moisture diffusivity in m^2/s and L is the thickness of slice (m).

Only the first term of equation (1) is used for long drying times (Lopez et al, 2000)

$$MR = \frac{8}{\pi^2} \exp\left[-\frac{\pi^2 Dt}{4L^2} \right] \quad (2)$$

The slope (K_0) in calculated by plotting In MR versus time according to Eq (3)

$$K_0 = \frac{\pi^2 D}{4L^2} \quad (3)$$

2.2 Determination of Activation Energy

The diffusivity coefficient at different temperatures is often found to be well predicted by the Arrhenius equation given by equation (4) as follows:

$$D_{eff} = \frac{DoeEa}{Rg(T + 273.15)} \quad (4)$$

Where, D_{eff} is the effective diffusivity coefficient m^2/s, D_o is the maximum diffusion coefficient (at infinite temperature), E_a is the activation energy for diffusion (KJ/mol), T is the temperature (^oC) and R_g is the gas constant.

Linearization the equation gives:

$$\ln Deff = -\left[\frac{1}{Rg(T+273.15)} Ea\right] + \ln Do \qquad (5)$$

D_o and E_a were obtained by plotting in D_{eff} against $-\left(\frac{1}{R_g\,(T+273.15)}\right)$

3.0 METHODOLOGY

Tomato seeds were purchased from the Nigeria Seed Services, Ibadan to ascertain its genetic purity and planted at the Osun State College of Education, Ilesa teaching and research farm. The tomato fruits were sorted for visual colour (completely red), size and physical damage. Osmotic solutions were prepared by mixing a blend of 45g/15g of sucrose/Nacl with 100 ml of distilled water to obtain a brix of 60 i.e. (60g of solute in 100ml of distilled water. Ibadan-local variety previously pretreated in 45/15/50 osmotic solution and untreated (fresh) samples were dried in the oven at 40, 45 and 50^oC until equilibrium weights were attained. Tomato samples (16 g each) were placed in 250 ml beakers, containing 160g of osmotic solution. The excess osmotic solution (fruit to solution ratio of 1:10) was used to limit concentration changes due to uptake of water from the tomato and loss of solute to the fruit. The samples were then immersed in a water bath and agitated to maintain a uniform temperature not more than $\pm 1^0C$ for the three temperature levels of 40, 45 and 50^oC. Samples were removed from the osmotic solution every 30 minutes until equilibrium was reached. Fruits were drained and the excess of solution at the surface was removed with absorbent paper (To eliminate posterior weight) and weighed using a top loading sensitive electronic balance (Mettler, P163). The water loss and solid gain were determined by gravimetric measurement and all determinations were conducted in triplicate.

The solid gain represents the amount of solid that diffuses from the osmotic solution into the tomato less the solid of the tomato that is lost to the solution. The values of water loss (WL) and solid gain (SG) have been presented by Mujica-Paz *et al*. (2003) and modified by Agarry *et al. (*2008) as;

$$WL = \frac{(Mo - mo) - (Mt - mt)}{Mo} \qquad (6)$$

$$SG = \frac{mt - mo}{Mo} \qquad (7)$$

Where, Mo is the initial weight of fresh tomato, mo is the dry mass of fresh tomato, Mt is the mass of tomato after time t of osmotic treatment and mo is the dry mass of tomato after time t of osmotic treatment .

Drying kinetics were compared using five existing models that describes the thin layer drying of high moisture products. The models used were: Exponential (Newton) model, Henderson and Pabis model, Page model, the modified page model and Logarithmic model. These were used to determine the activation energy and the effective coefficient of moisture diffusivity.

These osmotically treated samples were then subjected to oven drying at 40, 45 and 50^{o}C while untreated samples were also subjected to the same drying temperature in an oven that was previously run on a no-load mode for 30 min and the results were used to find the moisture ratio at different temperatures. The moisture ratio, MR (the ratio of free water still to be removed at time t to the total free water initially available in the food) was obtained by using the equation below (as given by Nieto *et. al*. 2001)

$$MR = \frac{Mt - Me}{Mo - Me} \qquad (8)$$

Where, Mt is the moisture content of tomato slab after time, t.

Me is the moisture content of tomato slab at equilibrium (gH_2O/g dry solid)

Mo is the moisture content of tomato slab prior to osmotic dehydration (g H_2O/g dry solid)

The drying time was thereafter plotted against time. The moisture Ratio, MR was also plotted against time at the different drying temperatures. Similarly, un-osmosized samples of tomatoes were also dried at the varying temperature of 40, 45 and 50^{o}C and weights were also taken at 30 min. interval until constant weights were obtained. The drying rate against time graph at the three temperatures and the MR plot against Time were further used for the drying kinetics. Simulation of results was done and fitted into five existing models viz: (Exponential (Newton) model, Henderson and Pabis model, Page model, Modified Page model and the Logarithmic model (Table 1) to predict mass transfer in the samples.

The initial parameter estimates were obtained by linearization of the models through logarithmic transformation and application of linear regression analysis. The least-squares estimates or coefficients of the terms were used as initial parameter estimates in the non-linear regression procedure. Model parameters were estimated by taking the moisture ratio (MR) to be the dependent variable. The Coefficient of determination (R^2), χ^2 and Root Mean Square Error (RMSE) were used as criteria for adequacy of fit. The best model describing the thin layer drying characteristics of tomato samples was chosen as the one with the highest R^2 and the least RMSE (Ozdemir and Devres, 1999; Doymaz *et al*., 2004; Ertekin and Yaldiz, 2004).

The experimental drying data for the determination of effective diffusivity coefficient (Deff) were interpreted using Fick's second law for spherical bodies according to Geankoplis (1983). This is because the shape of the seeds are closer to being spherical than the commonly used flat object (slab assumption). The diffusivity coefficient (Deff) was obtained from the equation for spherical bodies and the moisture diffusivity coefficient (Deff) was calculated at different temperatures using the slope derived from the linear regression of ln. (MR) against time data.

The effective radius (R) was calculated using the Aseogwu equation. The activation energy is a measure of the temperature sensitivity of Deff and it is the energy needed to initiate the moisture diffusion within the seed. It was obtained by linearising Equation (5)

4.0 RESULT AND DISCUSSION

Tables 2 to 9 show the results of the fitting statistics of various thin layer models at different drying temperatures

The result of the fitted models of treated samples at 40°C showed that the exponential, Henderson & Page and the Logarithmic model shared the same level of fit and the best fit . Page and Modified Page also shared the same level of fit, at 45°C, the exponential model had the best fit compared to others. Henderson and Pabis model and the Logarithmic model shared the same level of fit. Page and Modified Page also shared the same level of fit while at 50°C, Results showed that the Henderson and Pabis had the best fit. Exponential, Page and the Logarithmic model shared the same level of fit While modified Page have the lowest fit.

The result of the fitted models of untreated samples at 40°C showed that the Page model had the best fit. Exponential, Henderson & Page and the Logarithmic model shared the same level of fit, at 45°C, the Page and Modified Page have the best fit (Same level of fit). Exponential, Henderson and Page and the Logarithmic models shared the same level of fit, while models fitted at 50°C showed that the Modified Page have the best fit. Exponential, Henderson and Pabis and the Logarithmic models shared the same level of fit.

At different temperatures, different models fit in for the treated and untreated samples. The Exponential model fitted at 40 and 45°C with R^2 value range of 0.8291-0.8981 and 0.9352-0.981 for treated, 0.9453-0.9829 and 0.8281-0.9224 for untreated tomato having the best fit in Page and Modified Page and RMSE value range of 0.07966-0.10089,0.0464-0.364 (treated) and 0.0301-0.0538 (untreated) . While at 50⁰C, R^2 value ranged between 0.8461-0.8981 (treated) and 0.8281-0.9224 (untreated), with RMSE value of 0.07984-0.09659 and 0.0778-0.1008. Henderson and Pabis model gave the best fit for osmotically pretreated tomato and the Modified page fitting in for the untreated tomato. Calculated value of effective moisture diffusivity varied from 1.17-3.51x10^{-8} and 1.25-3.13x10^{-8} and the value of activation energy varied from a minimum of 46.81 to 52.61 to KJ/mol in treated and untreated tomato and R^2 value range of 0.977 to 0.919. It is obvious that the effective distribution coefficient in the samples dried at different temperatures (40, 45 and 50⁰C) varied between 1.17055 x 10^{-8} at 40⁰C, 2.34111 x 10^{-8} at 45⁰C and 3.51166 x 10^{-8}m^2/s. For osmotically pretreated sample to 1.25194 x 10^{-8} at 40⁰C, 2.50389 x 10^{-8} at 45⁰C and 3.12986 x 10^{-8} m^2/s at 50⁰C for untreated sample.

It can however be noted that the minimum effective coefficient moisture diffusivity (Deff) is in the lowest temperature (40⁰C). While the maximum Deff is in the highest drying temperature (50⁰C). However, the overall effective coefficient moisture diffusivity rate of food product observed was in 10^{-8}m^2/s for both the treated and the untreated tomato and this does not agree with the findings of Bablis and Belessiotis 2011.

A good understanding of the process mass transfer kinetics is of importance for a rational application of osmotic dehydration in fruits, obtaining efficient treatments and specific product formulations. The overall effective coefficient moisture diffusivity rate for food product has been assumed to change in the range of 10^{-11} to 10^{-9}. (Aghbashlo *et al.*, 2005)

Results indicated that there is a direct relationship between temperature and the effective spread, which shows that increase in temperature led to increase in the effective distribution coefficient. Temperature of 50°C has the highest value. Using the Arrhenius relationship earlier stated, the dependence of effective coefficient of moisture diffusivity to temperature was clearly described. Amplitude changes of effective coefficient of moisture diffusivity for tomato increased from 1.17 to 3.51 x 10^{-8} m^2/s in the temperature range of 40 to 50°C for treated and 1.25 to 3.13 x 10^{-8} m^2/s also in the same temperature range for untreated tomato.

The effective coefficients of moisture diffusivity increase with increase in drying temperature as observed by Garavand *et al.* (2011). In this study, the drying of tomato was only in the falling rate period and this implies that the moisture removal from the product was predominantly governed by diffusion phenomenon.

Findings from this study indicated that there is a direct relationship between temperature and the effective spread, which depicts that increase in temperature leads to increase in the effective distribution coefficient and this agrees with the findings of other researchers. Temperature of 50°C has the highest value of Deff. in direct humidity and intake speed conditions. Using the Arrhenius relationship, the dependence of effective coefficient moisture diffusivity to temperature was described correctly. Activation energy and constant effective coefficient diffusivity were calculated from the slope of Arrhenius (Ln (Deff) against 1/Tabs) are shown in tables 4.11 and 4.12. Changes of effective coefficient moisture diffusivity for tomato were gained from 1.17 x 10^{-8} to 3.52 x 10^{-8} in the temperature range of 40 to 50°C for osmotically pretreated samples local variety and 1.25 x 10^{-8} to 3.12 x 10^{-8} m^2/s in the same temperature range for untreated tomato.

Diffusivity constant value of 3.96 and 3.85 x10^{-8} m^2/s were obtained for treated and untreated samples. While the activation energy and R^2 value is higher in osmotically pretreated sample (52.61 KJ/mol) than untreated samples of tomato with activation energy value of (46.81 KJ/mol) and R^2 value of 0.92.

The effect of temperature on the diffusivity was expressed by the Arrhenius equation, where logarithm of the diffusivity exhibited a linear relationship against the reciprocal of the absolute temperature (R^2 = 0.98 (for treated tomato) and R^2 = 0.92 for untreated tomato) as can be observed in Figures 7 and 8.

5.0 CONCLUSIONS

- All the models used fitted but the Henderson and Pabis fitted best for osmotically pretreated tomato and modified page for untreated/fresh tomato as models with the highest values of X^2 and R^2 and the least RMSE. (These three were the criteria used to determine the degree of fitness of the models.)
- Of the entire five thin layer models used, the page and the modified page fitted in best.
- The present study has shown that the proposed empirical models was able to describe mass transfer process during osmotic dehydration of tomato as the values calculated using the proposed empirical models were in good agreement with the experimental data.
- Effective moisture diffusivity increases with increase in drying air temperature and coefficient of effective diffusion was found to be the least in air temperature of 40°C.
- Different models fitted in for the treated and untreated samples at different temperatures

6.0 RECOMMENDATIONS

This paper therefore recommends that a drying temperature of 50°C is best for effective spread and hence a high coefficient of moisture diffusivity. Also tomato should be pretreated osmotically to reduce the activation energy. However, further work should be done on the drying temperature limit that will not negatively affect the moisture diffusivity and the activation energy of pretreated tomato.

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Table 1: MATHEMATICAL MODELS USED FOR DRYING CHARACTERISTICS

MODEL	Model Equation
Exponential (Newton)	MR = exp (-kt)
Henderson and Pabis	MR = a.exp (-kt)
Page	MR = exp (-kt^n)
Modified Page	MR = exp [-(kt)^n]
Logarithmic	MR = a. exp (-kt)+c

Source: Akpinar and Bicer (2006)

Table 2: Results of the fitting statistics of various thin layer models at 40^{o}C drying temperature

Model no	Model name	Coefficients and constants	R^2	χ^2	RMSE
I	Exponential	k = 0.001	0.8981	380.02	0.07966
II	Henderson & Pabis	k = 0.001, a = 1.0023	0.8981	380.02	0.07984
III	Page	k = 0.001462, n = 0.980	0.8260	199.37	0.8260
IV	Modified page	k = 0.00128, n = 0.980	0.8219	199.40	0.10089
V	Logarithmic	k= 0.001, a = 1.0023, c = 0.00085	0.8981	380.02	0.07984

Table 3: Results of the fitting statistics of various thin layer models at 45^{o}C drying temperature

Model no	Model name	Coefficients and constants	R^2	χ^2	RMSE
I	Exponential	K = 0.002	0.981	2706.82	0.364
II	Henderson & Pabis	A = 1.002305, k = 0.002	0.9595	1233.91	0.0464
III	Page	K = 0.004009, n = 0.943	0.9352	735.88	0.06351
IV	Modified page	K = 0.002871524, n = 0.943	0.9352	735.49	0.06353
V	Logarithm	A= 1.002305, k = 0.002, c = 0.00189	0.9595	1233.91	0.04652

Table 4: Results of the fitting statistics of various thin layer models at 50°C drying temperature

Model no	Model name	Coefficients and constants	R^2	χ^2	RMSE
I	Exponential	K = 0.003	0.8664	214.04	0.0924
II	Henderson and Pabis	K = 0.003, a = 1.0225652	0.8981	380.02	0.0798
III	Page	K = 0.003475362, N = 0.987	0.8624	214.04	0.0945
IV	Modified page	K = 0.003225611, N = 0.987	0.8461	187.87	0.0966
V	Logarithmic	K = 0.003,a = 1.0225652, c = 0.00275	0.8624	214.04	0.0948

Table 5: **Results of the fitting statistics of various thin layer models at 40°C drying temperature of untreated local tomato**

Model no	Model name	Coefficients and constants	R^2	χ^2	RMSE
I	Exponential	K = 0.001	0.9453	881.61	0.0511
II	Henderson and Pabis	k=0.001,a = 1.04697	0.9453	881.61	0.0538
III	Page	k=0.000121, n= 1.274	0.9829	2938.62	0.0301
IV	Modified page	k = 0.000844, n = 1.274	0.9828	2912.56	0.0303
V	Logarithmic	K=0.001,a=1.04691, c = 0.00095	0.9453	881.61	0.0508

Table 6- Results of the fitting statistics of various thin layer models at 45°C drying temperature of untreated local tomato

Model no	Model name	Coefficients and constants	R^2	χ^2	RMSE
I	Exponential	k = 0.002	0.8281	246.71	0.1006
II	Henderson& Pabis	k=0.002, a = 1.02376	0.8281	246.71	0.1030
III	Page	k = 0.000179, n = 1.312	0.9224	607.28	0.0778
IV	Modified page	k = 0.001393, n = 1.312	0.9224	607.24	0.0778
V	Logarithmic	k=0.001,a= 1.04691, c = 0.00095	0.8281	246.71	0.1030

Table 7 - Results of the fitting statistics of various thin layer models at 50°C drying temperature of untreated local tomato

Model no	Model name	Coefficients and constants	R^2	χ^2	RMSE
I	Exponential	k = 0.002	0.8281	246.69	0.1006
II	Henderson and Pabis	k = 0.002, a = 1.0023	0.8283	246.99	0.1008
III	Page	k = 0.000244, n = 1.286	0.8963	441.66.	0.0899
IV	Modified page	k = 0.001553, n = 1.286	0.9224	607.24	0.0778
V	Logarithmic	k = 0.002, a = 1.0023, c = 0.1245	0.8321	248.84	0.0997

Table 8: Estimated effective moisture diffusivity at different temperature of drying for osmotically pre-treated tomato

	Diffusion	Coefficient	$10^{-8}(m^2/s)$
	40°C	45°C	50°C
Pre-treated tomato	1.17055	2.34111	3.51166
Untreated tomato	1.25194	2.50389	3.12986

Table 9: Estimated activation energy and moisture diffusivity constant at different temperatures

	Diffusion	**Coefficient**	**$10^{-8}(m^2/s)$**
	Do (m^2/s)	Ea (KJmol)	R^2
Pre-treated	3.963.58	52.61	0.977
Untreated	3.846118	46.81	0.919

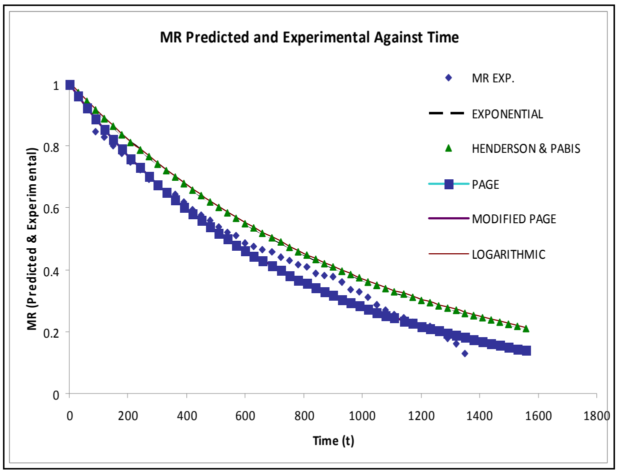

FIG.1: $MR_{EXP. \& PRE.}$ AGAINST TIME FOR TREATED TOMATO AT 45/15/50 DRIED AT 40°C

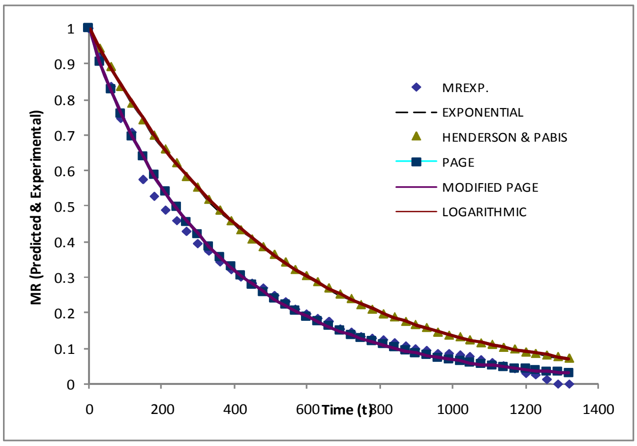

FIG.2: $MR_{EXP. \& PRE.}$ AGAINST TIME FOR TREATED TOMATO AT 45/15/50 DRIED AT 45°C

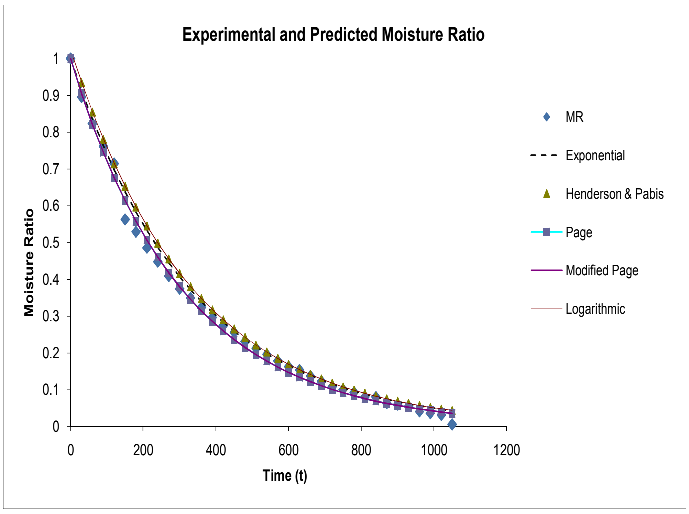

FIG. 3: $MR_{EXP. \& PRE.}$ AGAINST TIME FOR TREATED TOMATO AT 45/15/50 DRIED AT 50°C

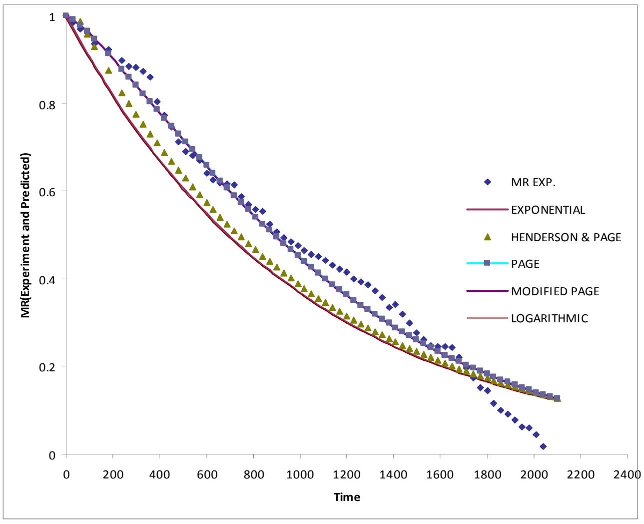

FIG. 4: $MR_{EXP. \& PRE.}$ AGAINST TIME FOR UNTREATED TOMATO DRIED AT 40°C

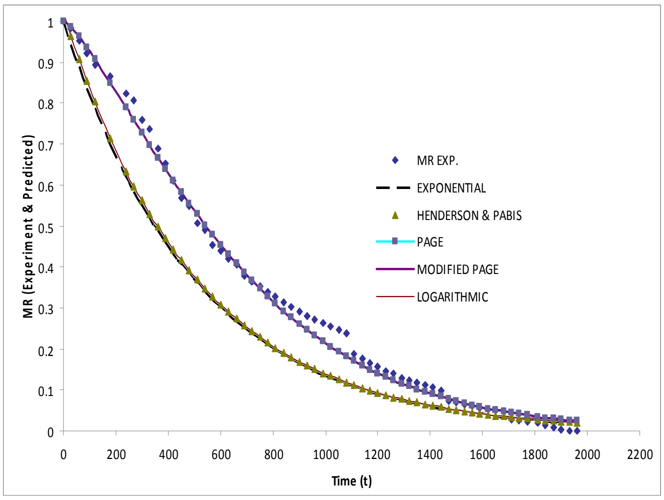

FIG. 5: $MR_{EXP. \& PRE.}$ AGAINST TIME FOR UNTREATED TOMATO DRIED AT 45°C

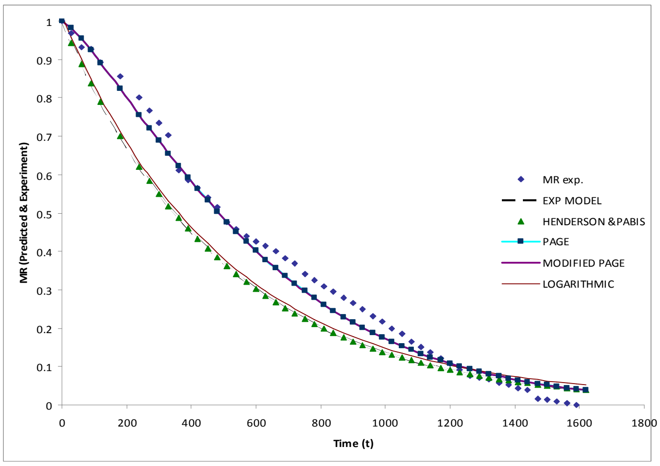

FIG. 6: $MR_{EXP. \& PRE.}$ AGAINST TIME FOR UNTREATED TOMATO DRIED AT 50°C

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