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Drying kinetics and moisture diffusivity study of ripe Jackfruit

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Abstract

This study presents the mathematical model of thin layer drying behavior of ripe jackfruit *(Artocarpus heterophyllus)*. The experiment was conducted at four different temperatures i.e. 50, 60, 70 and 80°C using a tray dryer. At varying temperature the dry basis moisture content values were subsequently used to fit 14 different thin-layer drying empirical models and among all, Middilli et al. model was found to be most suitable model representing the drying behavior of jackfruit. The constants and coefficients of Middilli et al. model were correlated with temperature and an equation was developed to describe the drying kinetics of jackfruit at a given time-temperature combination. Fick's second law of diffusion was used to characterize the effective diffusion of water molecules from the jackfruit pulp during the falling rate period drying process. The value of effective moisture diffusivity was found to increase with increase in temperature. From the analysis maximum diffusivity of 4.56 x 10^{-10} m²/sec was obtained at 80°C and minimum diffusivity of 1.264 x 10^{-10} m²/sec was obtained at 50°C. The effective moisture diffusivity was correlated with temperature by Arrhenius equation. The activation energy which is an indicator of minimum energy required to remove moisture from a solid matrix was found to be 40.846 kJ/mol. The pre exponential factor of Arrhenius equation was found to be 5 x 10^{-4} m²/s.

Introduction

Ripe jackfruit *(Artocarpus heterophyllus)* is mainly known for its consumption as a fruit or in the form of a dessert. It is native to parts of South and Southeast Asia, and is believed to have originated in the southwestern rain forests of India, in present-day Kerala, coastal Karnataka and Maharashtra. The flesh of the fruit is starchy, fibrous with sweet taste and exotic flavour. The carbohydrate content varies from 87.26 - 87.53% ; protein (2.93 - 4.39) % ; fat (2.51 - 2.75) % and fibre (2.54 - 3.86) % (Sonde *et al*., 1992). The bulbs (edible flakes) contain 7.5% sugar and a fair amount of Vitamin-A, Vitamin-C, potassium, protein, starch, calcium and thiamine (Burkill, 1997). Loaded with anti-ageing properties, the fruit slows down the degeneration of cells and makes the skin look young and supple (Swami *et al*., 2012). Since the fruit is not available all-round the year, drying acts as an alternative method of preservation for fruits (Mwithiga *et al*., 2005).

Drying of biological materials is a complicated process involving simultaneous, coupled heat and mass transfer phenomena occurring inside the material (Yilbas *et al*., 2003). It intends to enhance the storage life of the product for a longer period at room temperature and also brings about substantial reduction in weight and volume which minimizes the costs involved in packaging, storage, and distribution of the products (Potter and Hotchkiss, 1995). Drying of food is carried out either by sun drying method or by the mechanical means such as hot air dryers (Akpinar and Bicer, 2008). In tray dryers, the food is spread out in thin layers on trays where drying takes place. Heating may be by an air current sweeping across the trays, by conduction from heated trays or heated shelves on which the trays lie, or by radiation from heated surfaces (Earle, 1983). Most tray dryers are heated by blowing hot air.

The knowledge of effective moisture diffusivity is necessary for designing the mass transfer processes and activation energy describes the relative ease of moisture migration within the product. The literature highlights only a few studies on different properties and value-added products of ripe Jackfruit (Zuniga *et al*., 2006; Chien *et al*., 2008; Chong *et al*., 2008; Saxena *et al*., 2012). Therefore, to bridge the existing knowledge gap, the present study was undertaken to (i) investigate the effect of temperatures on the drying kinetics and (ii) model the drying data through

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14 different mathematical models and develop one as a function of air temperature (iii) correlate the effective moisture diffusivities with temperatures by Arrhenius-type equation.

Materials and Method

Experimental procedure

Jackfruit (*Artocarpus heterophyllus*, unripe) was purchased from the local markets (Tezpur, Assam) and was allowed to ripen under atmospheric conditions for a week. Optimum ripening is characterized by the fresh fruit aroma development and softening of the hard outer cover (pericarp). The drying behavior of ripe jackfruit was studied using a laboratory tray dryer (Model No.IK-112, IKON Instruments, New Delhi). The dryer is capable of providing a temperature range from 30 to 150°C. Drying kinetics is a function of temperature and, therefore, to study the effect of temperature, the experiments were conducted at four air temperatures of 50, 60, 70 and 80°C.The bulbs from the fruit were picked up and extracted from the seeds manually. The pulp, thus, obtained was spread in thin layers of approximately 3 mm thickness (Kaya *et al*., 2007a; Akpinar and Bicer, 2008) to allow full exposure of hot air in the drying chamber. During the drying process, the weight of the samples was recorded periodically at intervals of 30 minutes. The experiment was continued till it attained equilibrium moisture content. The moisture content of the sample was calculated by the oven drying method (Ranganna, 1986). In this process, about 5 g of the sample was weighed and kept in the oven at 105°C for 24 hours till it attained constant weight difference. The initial moisture content was calculated to be 77.56 % (wb).

Data analysis

The best fit model, that is used to describe the variation in the drying curves of the samples in the best possible way, is decided by the reduced chi-square (χ^2) and R square (R^2) values (Yaldiz and Ertekin, 2001; Menges and Ertekin, 2006; Goyal *et al*., 2007). Reduced chi-square is the mean square of the deviations between the experimental and calculated values of the models and was used to determine the goodness of fit. The model which has the minimum chi-square value and maximum R^2 value is regarded as the best model (Akpinar, 2006; Kaya *et al*., 2007a).

Many researchers have developed various semi-theoretical and empirical models for the study of thin layer drying. The Moisture Ratio (MR) was calculated at different temperatures and was fitted to the 14 different models (as listed in Table 1) generally used for majority of fruits and vegetables with diffusivity lying in the range of 10^{-9} to 10^{-11} m^2/s, by using non linear curve fitting tool of the Software Origin 8.5.

The MR can be expressed as presented in Eq. (1).

$$MR = \frac{Mt - Me}{Mo - Me} \quad (1)$$

Where, MR= Moisture Ratio, M_t= Moisture content at any time (kg water/ kg dry matter), M_o= Initial moisture content (kg water/ kg dry matter), M_e= Equilibrium moisture content (kg water/ kg dry matter).

As the equilibrium moisture content is relatively small i.e. (0.2 - 0.3) kg moisture/kg dry matter (db) in the experimental temperature range, MR term can be written in simplified from as (M_t-M_e / M_o-M_e) to (M_t/ M_o) and presented as shown in Eq. (2) (Yaldiz *et al.*, 2001; Midilli and Kucuk, 2003; Togrul and Pehlivan, 2004; Meziane 2011).

$$\frac{M_t}{M_0} \quad (2)$$

For most biological materials, Fick's second law of diffusion has been widely used to describe the drying process during the falling rate period (Tulek, 2011).The phenomenon of diffusion that occurs during involves surface diffusion, molecular diffusion and the combination of all these yields effective diffusivity. In thin layer drying, for different drying air temperatures, the curves of moisture ratio versus drying time are calculated based on Fick's second law of diffusion to estimate the effective moisture diffusivity. The Fick's second law of diffusion is shown in Eq. (3).

$$\delta M/\delta t = D\, \delta^2 M/\delta x^2 \quad (3)$$

Where, D = diffusivity (m^2/sec); $\delta M/\delta t$ = moisture content (db) per unit time (sec) and x = thickness (m).

By considering Jackfruit pulp as a slab having moisture distributed uniformly at a concentration M_0 and diffusion taking place only in the X direction, the following mathematical equations could be deduced (Eq. 4-7).

Initial conditions (t = 0):

$$M = M_0 \qquad 0 \le X < L \quad (4)$$

Boundary conditions (t > 0):

$$\left.\frac{\partial M}{\partial X}\right|_{X=0} = 0 \quad (5)$$

$$M = 0 \qquad X = L \quad (6)$$

Table 1. Thin layer drying curve models generally applied to the fruits and vegetables

Sl. No.	Model Name	Model	References
1	Newton	$MR = \exp(-kt)$	Westerman *et al*. (1973)
2	Page	$MR = \exp(-kt^n)$	Page (1949)
3	Modified page	$MR = \exp[-(kt)^n]$	Yaldiz *et al*. (2001)
4	Henderson and Pabis	$MR = a \exp(-kt)$	Henderson and Papis (1961)
5	Logarithmic	$MR = a \exp(-kt) + c$	Yaldiz and Ertekin (2001)
6	Two term	$MR = a \exp(-k_o t) + b \exp(-k_1 t)$	Henderson (1974), Rahman *et al*. (1998)
7	Two term exponential	$MR = a \exp(-kt) + (1-a)\exp(-kat)$	Yaldiz *et al*. (2001)
8	Wang and Singh	$MR = Mo + at + bt^2$	Ozdemir and Devres (1999)
9	Approximation of diffusion	$MR = a \exp(-kt) + (1-a)\exp(-kbt)$	Yaldiz and Ertekin (2001)
10	Verma et al.	$MR = a \exp(-kt) + (1 - a)\exp(-gt)$	Verma *et al*. (1985)
11	Modified Henderson and Pabis	$MR = a \exp(-kt) + b \exp(-gt) + c \exp(-ht)$	Karathanos and Belessiotis (1999)
12	Aghabashlo model	$MR = \exp(-k_1 t / 1+k_2 t)$	Aghabashlo *et al*. (2008)
13	Weibull	$MR = \exp((t\ a)^b)$	Corzo *et al*. (2008)
14	Midilli et al.	$MR = a \exp(-kt^n) + bt$	Midilli *et al*. (2002)

By solving Eq. (3), considering the conditions expressed in Eq. (4) - (6), unsteady state diffusion equation for slab geometry given by Crank (1975).

$$MR = \sum_{n=0}^{n=\infty} \frac{8}{(2n+1)^2 \pi^2} e^{\left(-\frac{(2n+1)^2 \pi^2 D_e t}{4L^2}\right)} \quad (7)$$

On simplifying Eq. (7), the mathematical solution of this model is represented in Eqn. (8) -(10)

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

Neglecting the higher terms,

$$MR = \frac{8}{\pi^2} \exp\left[-\frac{\pi^2 D_e t}{4L^2}\right] \quad (9)$$

Taking natural logarithm on both sides establishes a straight line relationship between logarithm of Moisture ratio (ln MR) and time (t) as shown in Eq. (10)

$$\ln MR = \ln \frac{8}{\pi^2} - \frac{\pi^2 D_e t}{4L^2} \quad (10)$$

The diffusion coefficient is calculated by the method of slopes. From the slope of the plot of ln MR versus time at different temperatures effective moisture diffusivity is calculated by application of Eq. (11).

$$Slope = -\pi^2 D_e / 4L^2 \quad (11)$$

Where, n= positive integer, D= effective moisture transfer diffusion coefficient (m2/s), L=half thickness (m) of jackfruit slab, and t = time (s).

Computation of activation energy

The temperature dependency of diffusivity can be illustrated by Arrhenius type equation as shown in Eq. (6).

$$D_e = D_0 \exp\left(-\frac{E_a}{RT}\right) \quad (12)$$

Taking natural logarithm on both sides, it can be written in a linear form as y= mx+ c as below:

$$\ln D_e = \ln D_0 - \left(\frac{E_a}{RT}\right) \quad (13)$$

Or,

$$\ln D_e = -\frac{E_a}{R} \cdot \frac{1}{T} + \ln D_0 \quad (14)$$

Where, D_0 = effective moisture diffusivity at infinite temperature (m^2/s), E_a= activation energy for diffusion (kJ/mol), R= gas constant (8.314×10^{-3} kJ/mol), T= temperature (K)

Results and Discussion

The initial moisture content of jackfruit pulp was found to be 77.56 % (wb). The experiments were conducted at different drying temperatures of 50, 60, 70 and 80°C until equilibrium moisture was achieved.

Fitting drying kinetic models

The moisture content values for different temperatures were converted to moisture ratio expression and then plotted with time as shown in Figure 1. Within the temperature range used, it is

Table 2. Model constants at different temperatures with average R2 value and χ2 value

Model	Temp (°C)	Constants	R^2 value	R^2 value (avg)	χ^2 value
Newton	50	k= 0.00687	0.98937		
	60	k= 0.00846	0.97672		
	70	k= 0.01304	0.9913	0.9875	0.001265
	80	k= 0.02906	0.99261		
Page	50	k= 0.00263, n= 1.1854	0.99748		
	60	k= 0.00473,n= 1.16551	0.9785		
	70	k= 0.00611,n= 1.11688	0.99648	0.993	0.000718
	80	k=0.00827, n=1.083061	0.99963		
Modified page	50	k= 0.00666, n= 1.18962	0.99748		
	60	k= 0.00822, n= 1.17244	0.97851		
	70	k= 0.01266, n= 1.11016	0.99648	0.993	0.000718
	80	k= 0.03305,n= 0.98029	0.99963		
Henderson and Pabis	50	k= 0.00434, a= 0.77143	0.9908		
	60	k= 0.0085, a= 0.98752	0.97481		
	70	k= 0.01336, a=1.00454	0.99116	0.9872	0.001292
	80	k= 0.02872, a= 1.02663	0.99206		
Logarithmic	50	a= 1.11241,k= 0.00559, c= -0.13157	0.7873		
	60	a= 1.09949, k= 0.0062, c= -0.1006	0.7073		
	70	a= 1.09297,k= 0.01088, c= -0.08531	0.7687	0.7473	0.000493
	80	a= 0.97308, k= 0.03086, c= -0.06925	0.7258		
Two term	50	a= 0.5716, k_0=0.0071,b= 0.46972, k_1= 0.00714	0.98927		
	60	a= 0.5923, k_0=0.00855, b= 0.4358, k_1= 0.00911	0.96982		
	70	a= 0.61288, k_0= 0.01336, b= 0.41374, k_1= 0.01336	0.98762	0.9866	0.001404
	80	a= 0.62666, k_0= 0.05203, b= 0.37344, k_1= 0.01442	0.99981		
Two term exponential	50	a= 1.71397, k= 0.00983	0.99728		
	60	a= 1.28817, k= 0.00937	0.97937		
	70	a= 0.99861, k= 0.00911	0.91977	0.9734	0.003019
	80	a= 0.31189, k= 0.00803	0.99776		
Wang and Singh	50	M_0= 0.9706, a=-0.00467,b= 5.71716E-6	0.99626		
	60	M_0= 0.93738, a= -0.00528,b= 7.39463E-6	0.98899		
	70	M_0= 0.88244, a= -0.00884,b= 2.03665E-5	0.99083	0.9360	0.005446
	80	M_0= 0.82966, a= -0.00916, b=1.59559E-5	0.768		
Approximation of Diffusion	50	a= 1.02738, k= 0.00687, b= 0.9992	0.98773		
	60	a= 1.06411, k= 0.00608, b= 0.55848	0.98711		
	70	a=1.13693, k=0.01468, b=0.34406	0.99463	0.9923	0.000795
	80	a= 1.26667, k= 0.05201, b= 0.2771	0.99983		
Verma et al.	50	a= 1.17636, k= 0.00685, g= 0.00685	0.98773		
	60	a= 1.01182, k= 0.00855, g= 0.00817	0.97257	0.9871	0.001348
	70	a=0.70205, k=0.01302, g=0.01302	0.9884		
	80	a= 0.37334, k= 0.01441, g= 0.05201	0.99983		
Modified Henderson and Pabis	50	a= 0.04009,k= 0.00714, b= 0.18722, g= 0.00714, c= 0.59403,h= 0.00713	0.98712		
	60	a= 0.16838, k=0.00849, b= 0.37263, g= 0.0085, c= 0.35339, h= 0.0085	0.96221		
	70	a=0.41979, k=0.01335, b=0.56638, g=0.01336, c=0.29047, h=0.01335	0.97936	0.982	0.001895
	80	a= 0.46326, k=0.05202, b= 0.77343, g=0.05210, c= 0.21352, h= 0.05202	0.99974		
Aghabashlo model	50	k_1= -0.06382, k_0= -0.0731,	0.98861		
	60	k_1=-0.07241, k_0=-0.08086	0.97478		
	70	k_1=-0.07708, k_0=-0.09012	0.99006	0.9863	0.001385
	80	k_1=- 0.08246, k_0= -0.1754	0.99187		
Weibull	50	a= 150.20728, b= 1.18956	0.99748	0.9930	0.000718
	60	a=111.57647, b=1.12205	0.97851		
	70	a=78.99056, b=1.07012	0.99648		
	80	a= 30.26247, b=1.03032	0.99963		
Midilli et al.	50	a= 0.9994,k= 0.00398, n= 0.72584, b= -1.9848E-4	0.99967		
	60	a=0.999, k=0.00842, n=0.93041, b=-3.276E-4	0.9869		
	70	a=1.00031, k=0.00972, n=1.07093, b=-4.0260E-4	0.99746	0.996	0.000423
	80	a= 1.0028, k= 0.00874, n= 1.08763, b= -1.12866E-4	0.99955		

evident that the time taken to reach the equilibrium moisture content shared an inverse relationship with temperature. The 80°C treatment showed the highest drying rate curve as compared to the other three treatments. Similar results have been obtained by other researchers in relation to other fruits and vegetables namely (Togrul and Pehlivan 2003; Doymaz, 2004; Goyal *et al.*, 2007; Kaya *et al.*, 2007a; Chien *et al.*, 2008; Raquel *et al.*, 2011; Tulek, 2011). The effect of drying air temperature, drying time, initial and final moisture content on the drying constants have been studied by many researchers (Misra *et al.*, 1980; Temple *et al.*, 1999; Yaldiz *et al.*, 2001; Midilli and Kucuk, 2003; Akpinar and Bicer, 2008). In this study, the most convenient model was obtained by statistical analysis of 14 different drying kinetic models proposed by earlier authors and the corresponding rate constants, R^2 value, chi-square values are presented in Table 2. Based on the minimum chi-square value and maximum R^2 value, Midilli *et al.* model (Eq. 15) was found to be the best suitable model representing the drying kinetics of jackfruit pulp. The constants and coefficients of the Midilli *et al*. model were correlated with temperature as below:

$$MR = a \exp(-kt^n) + bt \quad (15)$$

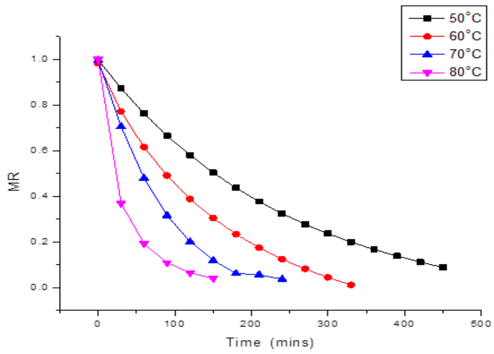

Figure 1. A plot of moisture ratio and time for experimental data of jackfruit at different temperatures

Where, a, k and n are constants and t is drying time in minutes and the relation with temperature is as follows.

$$n= 0.013T \quad (16)$$

$$k= -1x10^{-5}\, T^2 + 0.001T – 0.058 \quad (17)$$

$$b= -1x10\text{-}6\, T^2 + 0.12 \quad (18)$$

$$a=1.00 \quad (19)$$

The above correlations (Eq. 16 to 18) showed R^2 values of 0.929, 0.998 and 0.898 representing good fitting of the model parameters. Combing Eqn. 15 to 19 moisture ratio can be presented as shown in Eq. (20)

$$MR = \exp(-(-1x10^{-5}\, T^2 + 0.001T – 0.058)\, t^{0.013T}) +(-1x10^{-6}\, T^2 + 0.12)\, t \quad (20)$$

Table 3. Diffusivities of jackfruit pulp at different temperatures.

Temperature (°C)	Diffusivity (m^2/s)	R^2
50	1.264×10^{-10}	0.999
60	2.28×10^{-10}	0.984
70	3.55×10^{-10}	0.995
80	4.56×10^{-10}	0.966

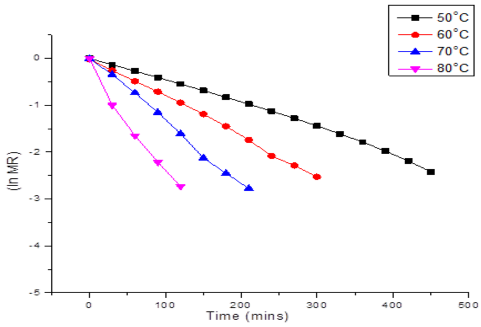

Figure 2. A plot of ln MR and time of jackfruit at different temperatures

Eq. (20) could be used to predict the moisture ratio at a definite time- temperature combination for the drying of jackfruit pulp. The R^2 value of the equation was found to be 0.946. A high value of correlation coefficient suggests that the predicted model is fitting well with the experimental data.

Effective Diffusivity

A curve was plotted between logarithm of moisture ratio versus time (min) for the different temperatures as illustrated (Figure 2) and the effective moisture diffusivity was calculated from the obtained slope. The values of ln MR were found to be in the negative range and similar results have been obtained by other researchers (Karathanos and Belessiotis, 1999; Sharma and Prasad, 2004; Akintunde and Ogunlakin, 2011). From the slope the estimated effective diffusivity was found to be in the range of 1.264×10^{-10} and 4.56×10^{-10} m^2/s at 50-80°C as shown in Table (3). The values of Deff were reported to vary between ($2.4 \times 10^{-10} – 6.22 \times 10^{-10}$) m^2/s for grapes at 50-70°C (Pahlavanzadeh *et al.*, 2001); (0.65×10^{-10} - 6.92×10^{-10}) m^2/s for quince at 35-55°C (Kaya *et al.*, 2007b); ($1.51 \times 10^{-10} – 5.32 \times 10^{-10}$) m^2/s for cactus pears at 40-70°C (Ruiz-Cebrera *et al.*, 2008); (3.32×10^{-10} – 9.0×10^{-10}) m^2/s for berberies at 50-70°C (Aghbashlo *et al.*, 2009). Further, a general trend of increase in the effective diffusion with increase in temperature was observed as obtained by many researchers for different fruits and vegetables (Doymaz, 2005; Akpinar, 2006; Kaya *et al.*, 2007a; Tulek, 2011; Radhika, *et al.*, 2011; Rayaguru and Routray, 2012). The effect of temperature on effective diffusivity is described using Arrhenius type equation. The values of effective diffusivity (De) at five temperatures were calculated from Eq. (5). ln-De was plotted with 1/T as shown in Figure 3. From the slope of the straight line, the activation energy Ea was estimated as 40.84 kJ/mol. The activation energy was consistent with the values obtained for the drying of pear (33.56 kJ/mol) (Ruiz-Cebrera *et al.*, 2008), fig (40.95 kJ/mol) (Xanthopoulos, 2009) and olive pomace (34.05 kJ/mol) (Meziane, 2011). The pre-exponential factor Do was obtained as 5×10^{-4} m^2/s and similar values have been obtained for other fruits and vegetables like grape seed (7.79×10^{-5} m^2/s) (Roberts *et al.*, 2008), cocoa (8.43×10^{-4} m^2/s) (Hii *et al.*, 2009) and gooseberries (5.53×10^{-4} m^2/s) (Vega-Galvez *et al.*, 2012). The correlation is represented as

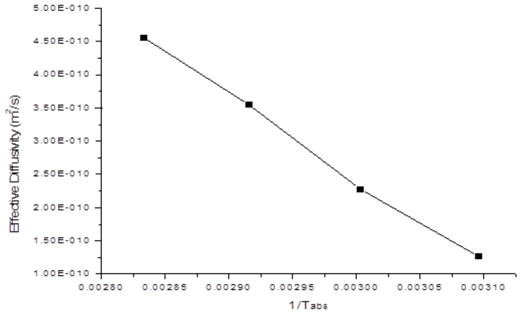

Figure 3. Variation of effective diffusivity with absolute temperature

$$De = 5\times10^{-4}\exp\left(-\frac{40.84}{RT}\right) \qquad (21)$$

The pre-exponential factor (D_o) in Arrhenius equation represents the diffusivity constant equivalent to the diffusivity at infinitely high temperature. The activation energy is the relative ease of moisture migration within the product and a lower value indicates high moisture diffusivity (Sharma and Prasad, 2004). Hence, in the present study about 40.84 kJ/mol of energy is required for the moisture diffusion and subsequent evaporation from the surface of the fruit.

Conclusion

Dried jackfruit powder is commercially manufactured in countries like Thailand, China, Vietnam, United Kingdom and Ukraine and is used for the production of ice-creams, falvored drinks, extracts etc. The present study shows the drying characteristics of jackfruit and the transfer processes during drying of jackfruit pulp to obtain quality dried jackfruit leather. The highest effective diffusion was found to be 4.56 x 10^{-10} at 80°C and the lowest was found to be 1.264 x10^{-10} at 50°C. The Activation energy was observed to be 40.84 kJ/mol. According to the results obtained, the Midilli *et al.* model adequately described the drying behavior of jackfruit. The values of constants were effectively correlated with temperature and the model will provide reliable predictions of the moisture distributions of jackfruit pulp at any instant of time during the drying process.

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