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Experimental and modelling study of thin layer drying kinetics of pellets millet flour

P. T. Bassene[1]*, S. Gaye[1], A. Talla[2], V. Sambou[1] and D. Azilinon[1]

[1]Laboratoire d'Energétique Appliquée (LEA), Ecole Supérieure Polytechnique (ESP), Université Cheikh Anta Diop (UCAD) BP 5085 Dakar-Fann, Dakar, Sénégal.
[2]Laboratoire d'Energétique d'Eau et de l'Environnement, Ecole Nationale Supérieure Polytechnique (ENSP), BP 8390, Yaoundé, Cameroun.

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The thin layer drying kinetics of pellets millet flour was studied at drying air temperatures of 40, 50 and 60°C using a constant air velocity of 1.0 m/s for pellets size 2.5 mm < d_1 < 3.15 mm and 5 mm < d_2 < 6.30 mm. Ten thin layer drying models were evaluated by fitting the experimental moisture data. The goodness of fit of each model was evaluated using the coefficient of determination (R^2) the reduced chi-square (χ^2) and root means square error (RMSE). Among these models, the modified Henderson and Pabis model is the best one to describe the behaviour in the drying of pellets millet. The effective diffusivity has been found to be varying between 4.4676 × 10^{-9} and 31.94 x 10^{-9} m^2/s and activation energy was 33.048 and 42.658 kJ/mol for pellets size of d_1 and d_2 respectively.

Key words: Thin layer drying, mathematical modelling, effective diffusivity, activation energy.

INTRODUCTION

Cereals are the main food in Sahel countries. There is a strong demand for grain products in both rural and urban areas. This sudden and unexpected interest for local cereals, once abandoned, is interesting in agriculture and agribusiness. It will finally allow finding enough opportunities for these local grains and thus reviving the cultivation of cereals including millet.

Drying process is one of the oldest methods of agricultural products preservation (Meziane, 2011). It is the most important process to preserve grains, crops and foods of all varieties. The removal of moisture prevents the growth and reproduction of microorganisms causing decay and minimises many of the moisture-mediated deterioration reactions. It brings about substantial reduction in weight and volume, minimising packing, storage and transportation costs and enables storability of the product under ambient temperatures (Doymaz, 2008).

In order to study the effects of the drying on the pellets millet it is necessary to have a deep knowledge of the mass transfer parameters and the drying kinetics. These characteristics are considered to be important for the design, simulation and optimization of the drying process by using mathematical modelling. The mathematical models have proved to be very useful in the new design and/or improvement of drying systems and analysis of mass transfer phenomena involved during drying (Akpinar et al., 2008).

Thus it is necessary to have an accurate model able to predict water removal rates and describe the drying performance of each product under the common conditions used in normal commercial relevant facilities (Flores et al., 2012). Therefore several researchers have investigated the drying kinetics of various agricultural

*Corresponding author. E-mail:pbassene@hotmail.com. Tel: +221 77 519 42 11.

products in order to evaluate different mathematical models for describing drying characteristics. Belghit and Bennis (2009) made experimental analysis of the drying kinetics of cork. Doymaz (2008) investigated the drying kinetics of strawberry. Drying kinetics of nopal *(Opuntia ficus-indica)* using three different methods and their effect on their mechanical properties were studies by Torres et al. (2008); Lertworasirikul (2008) investigated drying kinetics of semi-finished cassava crackers. Roberts et al.(2008) studied drying of grape seeds representing waste products from white wine processing *(Riesling)*, red wine processing *(Cab Franc)*, and juice processing *(Concord)*. Chong et al. (2008) investigated drying kinetics and product quality of dried chempedak. Recently, Folres et al. (2012) studied the drying kinetics of castor oil seeds during fluidized bed drying and Mezaine (2011) investigated drying kinetics of olive pomace in a fluidized dryer. Arumuganathan et al. (2009) studied the drying kinetics of milky mushroom slices in a fluidized bed dryer.

Thin layer drying equations describe the drying phenomena in a unified way, regardless the controlling mechanism. They have been used to estimate drying times of several products and to generalize drying curves. In the development of thin layer drying models for agricultural products, generally the moisture content of the material at any time after it has been subjected to a constant relative humidity and temperature condition is measured and correlated to the drying parameters (Akpinar et al., 2008). Many studies have recently focused on thin layer drying of agricultural products, fruits, and food materials. Thin layer drying characteristics of the finger millet *(Eluesine coracane)* were studied by Radhika et al. (2011) and in their work drying data were fitted to nine thin layer models. The logarithmic model was found to satisfactorily describe the drying kinetics of the finger millet. Shen et al. (2011) studied thin-layer drying kinetics and quality changes of sweet sorghum stalk for ethanol production as affected by drying temperature. The drying kinetics and the effects of drying temperature on the qualities of sweet sorghum stalk were investigated in their work. The results showed that the drying process could be well simulated by Wang and Singh's model. Mathematical modelling of the thin layer drying kinetics of *G. tournefortii* was investigated for both the microwave and open sun drying conditions by Evin (2011). The experimental moisture loss data were fitted to the 14 thin layer drying models and the logarithmic model was found to best describe the open sun drying kinetics of *G. tournefortii.*

Characteristics of Amelie and Brooks mangoes were experimentally determined using a solar dryer made up of four trays and used under weather conditions of fruit harvest period by Dissa et al. (2011). Direct solar drying curves were established, fitted using 10 mathematical models and simulated with a direct solar drying model.

Other investigators have successfully used thin layer equations to explain drying of several agricultural products. For example, Roselle (I) (Saeed et al, 2008); green pepper (Akpinar et al., 2008). However, in our knowledge, no information is available about thin layer drying of pellets millet. Therefore, in the present study, thin layer drying kinetic of pellets millets was investigated. The main objectives of the present work were:

1. To investigate the drying kinetics of pellets millet
2. To discuss the influence of temperature and size pellets on the drying kinetics
3. To fit the drying kinetics with ten mathematical models and select, the best model to represent accurately the drying kinetics of pellets millets
4. To evaluate the effective moisture diffusivity coefficients and activation energy.

MATERIALS AND METHODS

Sample preparation

Pellets are prepared from the flour of millet. The manufacture of the product consists of humidifying the flour then to roll it to the hand in order to obtain pellets the dimensions of which depend on the quality of the required product. Pellets sizes are obtained by using a sieves system and the initial moisture content is 0.602 kg water / kg dry matter for pellets 2.5 mm < d_1 < 3.15 mm and 0.628 kg water / kg dry matter for pellets 5 mm < d_2 < 6.30 mm.

Experimental apparatus

The experimental device used is a blower with air called vein of drying. It includes:

1. An axial fan driven by an engine;
2. A comprising low tension battery of resistance which makes it possible to heat the air;
3. A test vein of section 175 × 175 mm^2
4. A convergent at the entry of the sheath for better channelling the flow of air;
5. A power station of data acquisition connected to a microcomputer equipped with the software for the management of the power station, storage and the data processing;
6. A site to put the product to be dried;
7. An electronic balance of precision ± 0.1 g for monitoring the evolution of the mass in the course of drying;
8. An digital anemometer for the control rate of air flow in the test vein.

Experimental procedure

Initially, the desired conditions of drying, namely the temperature T, the relative humidity H_R and the air velocity have to be fixed. By varying fan tension, the air flow and temperature of drying was

Table 1. Thin layer mathematical models used to describe the drying kinetics of pellets.

N°	Model name	Model equation	Reference
1	Newton	$X_R = \exp(-kt)$	Shen et al. (2011), Evin (2011), Meziane (2011), Akpinar et al.(2008)
2	Page	$X_R = \exp(-kt^n)$	Flores et al. (2012) ,Radhika et al.(2011), Shen at al.(2011), Evin (2011), Meziane (2011) , Doymaz (2008) , Akpinar et al.(2008)
3	Henderson and Pabis	$X_R = a\exp(-kt)$	Flores et al. (2012) , Radhika et al. (2011), Shen et al. (2011), Evin (2011), Meziane (2011) , Doymaz (2008) , Akpinar et al. (2008)
4	Logarithmic	$X_R = a\exp(-kt) + c$	Radhika et al.(2011), Shen at al.(2011), Evin (2011), Meziane (2011), Doymaz (2008) , Akpinar et al.(2008)
5	Two-term	$X_R = a\exp(-k_0 t) + b\exp(-k_1 t)$	Flores et al.(2012) , Shen at al.(2011), Evin (2011), Meziane (2011), Akpinar et al.(2008)
6	Diffusion approach	$X_R = a\exp(-kt) + (1-a)\exp(-kbt)$	Flores et al. (2012) , Meziane (2011), Akpinar et al. (2008)
7	Modified Henderson and Pabis	$X_R = a\exp(-kt) + b\exp(-gt) + c\exp(-ht)$	Evin (2011), Meziane (2011), Akpinar et al. (2008)
8	Two term exponential	$X_R = a\exp(-kt) + (1-a)\exp(-kat)$	Shen at al.(2011), Evin (2011)
9	Wang and Sing	$X_R = 1 + at + bt^2$	Flores et al. (2012) , Radhika et al(2011), Shen et al. (2011), Meziane (2011), Doymaz (2008)
10	Midilli et al	$X_R = a\exp(-kt^n) + bt$	Evin (2011), Meziane (2011)

controlled during the whole operation. The fan makes the air pass through electrical resistances where it is heated. Once the experimental conditions are stable, the product is put on an aluminium plate placed in parallel to the hot air flow. To follow the product weight losses during drying, weight measurements were taken every five minutes. Drying procedure goes on until there is no weight change after three successive readings.

Mathematical modelling of the drying curves

Ten simplified drying models given in Table 1 have been used to describe the drying kinetics of pellets millet. In these models, X_R represents moisture ratio expressed by the following equation:

$$X_R = \frac{X_t - X_e}{X_0 - X_e} \quad (1)$$

Where X_t is the moisture content at t time, X_0 and X_e are respectively the initial and equilibrium moisture contents on dry basis. X_e is relatively small compared to X_t and X_0 (Meziane, 2011; Radhika et al., 2011; Evin, 2011; Dissa et al., 2011; Arumuganathan et al., 2009; Akpinar et al., 2008; Doymaz, 2007;). Thus, X_R can be simplified by $X_R = X_t / X_0$.

A non-linear regression analysis was performed using the Microsoft excel 2010 solver to fit the experimental data with chosen mathematical models. The statistical validity of the models was evaluated and compared by means of the coefficient of determination (R^2) the reduced chi-square (χ^2) and root means square error (RMSE). The best fit is defined by the highest value of R^2, and the lowest values of χ^2 and RMSE. These parameters can be calculated as follows:

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

$$\chi^2 = \frac{\sum_{i=1}^{N} \left(X_{R\exp,i} - X_{R\mathrm{pre},i} \right)^2}{N - p} \quad (3)$$

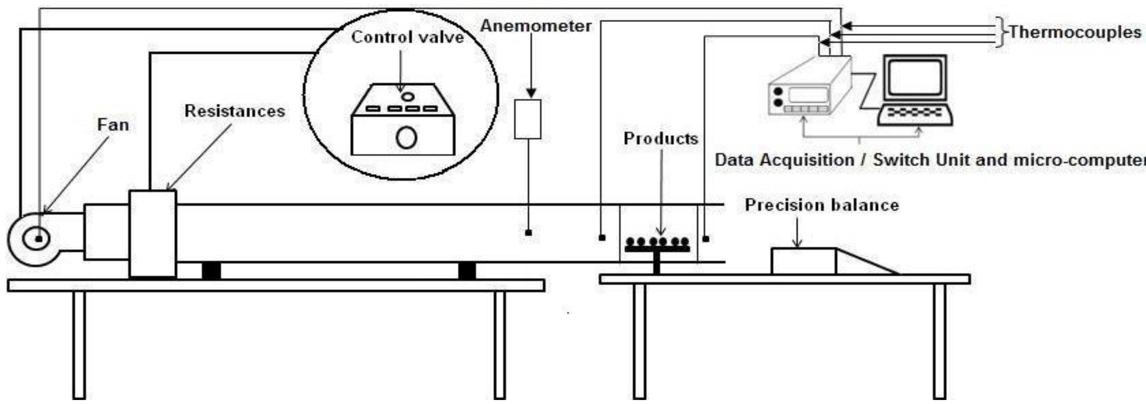

Figure 1. Schema of the thin layer drying apparatus used in the experiments.

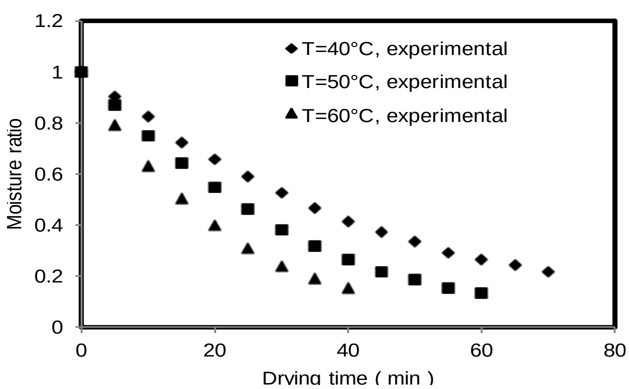

Figure 2. Effect of drying air temperature on moisture ratio for pellets size d_1.

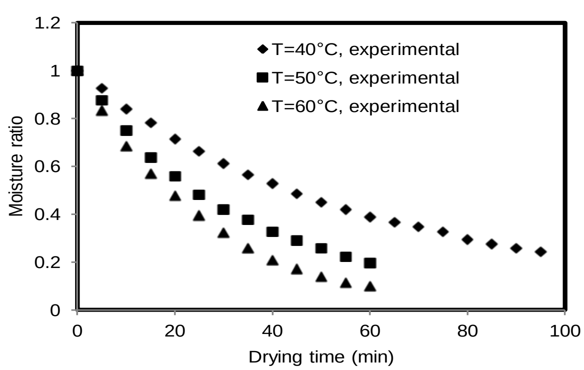

Figure 3. Effect of drying air temperature on moisture ratio for samples pellets size d_2.

Where $X_{Rexp,i}$ is the *i*th experimental moisture ratio, $X_{Rpre,i}$ is the *i*th predicted moisture ratio, *N* is the number of experimental data points, and *p* is the number of parameters in model (Dissa et al., 2011 ; Roberts et al., 2008; Doymaz , 2008).

RESULTS AND DISCUSSION

Effect of drying air temperature on moisture ratio of pellets millet

Pellets were dried in thin layer at the drying air temperature of 40, 50 and 60°C in a convective hot air dryer using a constant air velocity of 1.0 m/s (Figure 1). Figures 2 and 3 present the variations of moisture ratio depending on time for various used air temperatures and diameters 2.5 mm < d_1 < 3.15 mm and 5 mm < d_2 < 6.30 mm respectively.

Moisture ratio decreases continuously with drying time and a constant-rate period is not observed at none of the experiments. All the drying process occurred during the falling rate-drying period. This indicates that the process describing the drying behaviour of the pellets millet is governed by diffusion. In fact, an increase in drying air temperature not only modifies water activity but also influences the diffusion coefficient and to a lesser extends the vaporization enthalpy. Similar results related to behaviour or drying rate curves have also been reported in several drying studies of biological materials by Flores et al. (2012) for castor oil seeds *(Ricinus communis)*, Radhika et al. (2011) for Finger Millet *(Eluesine coracana)*, Shen et al. (2011) for sweet sorghum; Meziane (2011) for olive pomace, and Doymaz (2008) for strawberry.

Effect of the pellets size

The influence of the pellets size is highlighted by changing pellets size from one test to another. Figure 4 shows the effect of the pellets size on drying kinetics at various drying air temperatures. Decreasing pellets size increases moisture ratio and consequently drying time

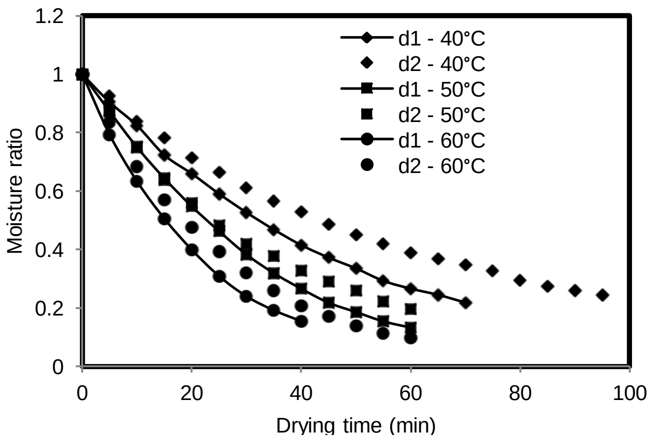

Figure 4. Effect of the pellets size on the kinetics of drying at various drying air temperatures.

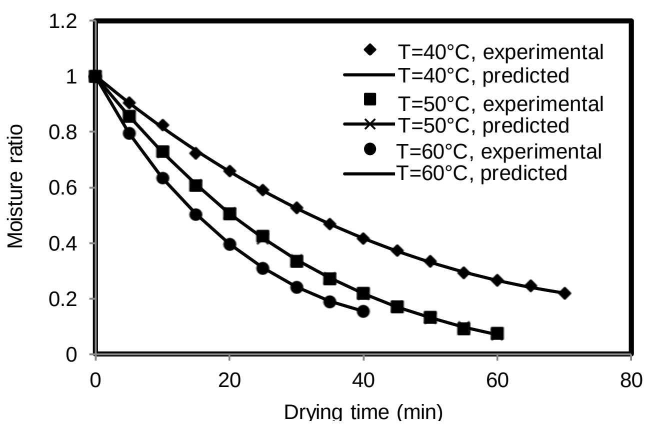

Figure 5. Variations of experimental and predicted moisture ratios by the Henderson and Pabis modified drying model with drying time for pellets size d_1 and V= 1 $m.s^{-1}$.

decreases. Decreasing pellets size accelerates the migration of water from inside to the surface of the product and a decrease of surface exchange increases heat transfer between the air and the product. Similar result has been noted by Madamba et al. (1995) for garlic slices.

Modelling of drying curves

The moisture ratio data obtained from the drying experiments were fitted to the 10 thin layer models listed in Table 1. The values of the coefficient of determination (R^2), the reduced chi-square (χ^2) and the root mean square error (RMSE) for different temperatures and pellets size determined by non-linear regression analysis

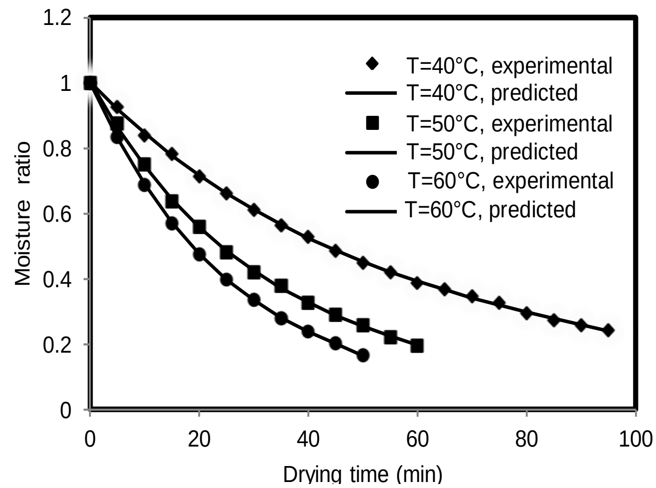

Figure 6. variations of experimental and predicted moisture ratios by the Henderson and Pabis modified drying model with drying time for pellets size d_2 and V= 1 $m.s^{-1}$.

are presented in Table 2. In all cases, the R^2 values for the models were greater than the acceptable R^2 value of 0.90, indicating a good fit (Doymaz, 2008). For all cases, Henderson and Pabis' modified model gives the best fit of experimental values since highest value of R^2 and lowest values of χ^2 and RMSE were obtained. Accordingly, the Henderson and Pabis' modified model can be selected as a suitable model to represent the thin layer drying behaviour of pellets millets. Coefficients and constants of the best drying model for different drying condition are given in Table 3. Figures 5 and 6 represent the variations of experimental moisture ratio and predicted moisture ratio by the Henderson and Pabis modified drying model for various drying air temperatures and for pellets size d_1.and d_2 respectively.

Estimation of effective diffusivities

It has been accepted that the drying characteristics of biological products in falling rate period could be described by Fick's diffusion equation (Flores et al., 2012; Shen et al., 2011; Doymaz, 2008). General series solution of Fick's second law in spherical coordinates, with the assumptions of moisture migration being by diffusion, negligible shrinkage, constant diffusion coefficients and temperature are given as follows (Crank, 1975; Doymaz, 2008; Flores et al., 2012):

$$X_R = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left[-n^2\frac{\pi^2 D_{eff}\,t}{r^2}\right] \qquad (4)$$

Where X_R is the moisture ratio, D_{eff} is the effective

Table 2. Statistical results obtained from 10 thin layer drying models.

Model	T°C	Pellets size					
		2.5 mm < d_1 < 3.5 mm			5 mm < d_2 < 6.30 mm		
		R^2	χ^2	RMSE	R^2	χ^2	RMSE
Newton	40	0.99969	0.000063	0.007642	0.99931	0.000121	0.010723
	50	0.99915	0.000292	0.016406	0.99932	0.000109	0.010049
	60	0.99987	0.000027	0.004872	0.99979	0.000053	0.006980
Page	40	0.99980	0.000027	0.004867	0.99975	0.000030	0.005180
	50	0.99992	0.000014	0.003477	0.99957	0.000065	0.007392
	60	0.99991	0.000018	0.003690	0.99988	0.000023	0.004371
Henderson and Pabis	40	0.99967	0.000045	0.006241	0.99915	0.000099	0.009428
	50	0.99891	0.000191	0.013279	0.99927	0.000111	0.009682
	60	0.99986	0.000028	0.004693	0.99976	0.000048	0.006372
Logarithmic	40	0.99973	0.000041	0.005707	0.99982	0.000021	0.004220
	50	0.99965	0.000069	0.007304	0.99976	0.000039	0.005453
	60	0.99991	0.000019	0.003603	0.99988	0.000024	0.004288
Two term	40	0.99974	0.000042	0.005573	0.99986	0.000018	0.003773
	50	0.99993	0.000020	0.003320	0.99977	0.000042	0.005369
	60	0.99992	0.000022	0.003475	0.99976	0.000059	0.006372
Diffusion approach	40	0.99983	0.000026	0.004522	0.99929	0.000146	0.011152
	50	0.99915	0.000292	0.015273	0.99975	0.000044	0.005806
	60	0.99987	0.000027	0.004872	0.99979	0.000063	0.006980
Modified Henderson and Pabis	40	0.99986	0.000027	0.004010	0.99987	0.000019	0.003647
	50	0.99993	0.000020	0.003288	0.99977	0.000052	0.005285
	60	0.99996	0.000016	0.002333	0.99994	0.000018	0.003133
Two term exponential	40	0.99976	0.000033	0.005362	0.99929	0.000138	0.011152
	50	0.99915	0.000318	0.016406	0.99974	0.000041	0.005866
	60	0.99987	0.000031	0.004872	0.99979	0.000058	0.006980
Wang and Singh	40	0.99982	0.000025	0.004671	0.99881	0.000167	0.012276
	50	0.99994	0.000011	0.003088	0.99854	0.000240	0.014254
	60	0.99945	0.000140	0.010433	0.99882	0.000367	0.015949
Midilli et al.	40	0.99985	0.000024	0.004188	0.99977	0.000027	0.004808
	50	0.99992	0.000016	0.003378	0.99975	0.000045	0.005588
	60	0.99991	0.000023	0.003585	0.99989	0.000025	0.004153

moisture diffusivity (m^2/s), r is the equivalent radius (m) and t is the time (s). For long drying time, Equation (4) could be further simplified to retain only the first term (Flores et al., 2012; Radhika et al., 2011). It could be rewritten in a logarithmic as shown in Equation (5). The effective moisture diffusivity was calculated from the slope of a straight line, plotting experimental data in terms of ln (X_R) versus drying time (Flores et al., 2012; Doymaz, 2008):

$$\ln X_R = \ln \frac{6}{\pi^2} - \left(\frac{\pi}{r}\right)^2 D_{eff}\, t \quad (5)$$

The effective moisture diffusivity was estimated by using

Table 3. Coefficients and constants of the Modified Henderson and Pabis model for different drying condition.

Pellets size	Model constants						
	T°C	k	a	b	c	g	h
2.5 mm < d_1 < 3.5 mm	40	0.0101	2.7666	-2.1394	0.3741	0.0022	-0.0089
	50	0.0449	3.1382	1.9919	-0.1454	0.0540	0.0455
	60	0.0441	1.0049	-0.3054	0.2983	0.0802	0.0807
5 mm < d_2 < 6.30 mm	40	0.0188	0.9399	0.758	-0.0131	-0.0107	-0.0237
	50	0.0349	0.8863	0.3135	-0.1935	-0.0075	-0.0123
	60	0.0372	1.0019	0.0025	-0.0034	-0.0965	-0.0916

Table 4. Effective diffusivity values of pellets millet.

Diameter		d_1			d_2	
T°C	40	50	60	40	50	60
$D_{eff} \times 10^{-9}$ ($m^2.s^{-1}$)	4.468	6.974	9.562	11.967	21.832	31.940

the method of slopes. From Equation (5), a plot of ln X_R versus time gives a straight line with a slope of:

$$\text{slope} = \frac{\pi^2 D_{eff}}{r^2} \qquad (6)$$

The effective diffusivity values for different temperature and pellets size are presented in Table 4. The effective diffusivities vary from 4.468 × 10^{-9} to 6.974 × 10^{-9} m^2/s for pellets size d_1 and from 11.967 × 10^{-9} to 31.940 × 10^{-9} m^2/s for pellets size d_2. Generally in the biological products, this value varies between 10^{-9} and 10^{-11} m^2/s (Doymaz, 2008; Flores et al., 2012; Madamba et al., 1995). The effective diffusivity increases when drying air temperature s and pellets size increase. These results are in agreement with those of Meziane (2011) for olive pomace.

Estimation of activation energies

The temperature dependence of effective moisture diffusivity can be described by an Arrhenius type relationship:

$$\text{Deff} = D_0 \exp\left(-\frac{E_a}{RT}\right) \qquad (7)$$

Where D_{eff} is the effective moisture diffusivity (m^2/s), D_0 is the pre-exponential factor equivalent to the diffusivity at a high temperature (m^2/s), E_a is the activation energy (kJ/mol), R is the universal gas constant (8,314 J/mol K), and T is absolute temperature (K). The activation energy E_a and the constant D_0 could be determined by plotting ln (D_{eff}) versus 1/T after linearization for Equation (7). The values of E_a calculated from the slope and intercept of each plot are given in Table 5. As it can be seen, the activated energy increases when pellets size increase. This observation is in agreement with those reported by Meziane (2011).

Similar activation energy values have been found by several authors, from 35.37 kJ/mol for finger millet (Radhika et al., 2011); in the range of 34.05 to 38.10 kJ for olive pomace (Meziane, 2011), and 41.41 kJ/mol for castor oil seeds *(Ricinus cumminis)* (Flores et al., 2012).

Conclusions

In this study, thin layer drying kinetics of pellets millet flour was studied at drying air temperatures of 40, 50 and 60°C.using a constant air velocity of 1.0 m/s for diameters 2.5 mm < d_1 < 3.15 mm and 5 mm < d_2 < 6.30 mm. The drying process takes place only in falling rate period. Drying air temperature and the pellets size are influencing factors to drying kinetics. Statistical results of ten mathematical models at different drying conditions showed that the modified Henderson and Pabis is the best model to describe the behaviour of pellets millet drying. The effective diffusivity varies from 4.468 × 10^{-9} to 9.562 × 10^{-9} m^2/s for d_1 and from 11.967 × 10^{-9} to 31.940× 10^{-9} m^2/s for d_2. It increases when drying air

Table 5. Arrhenius parameters for the drying kinetics of granulated of flour of millet.

D(mm)	d_1	d_2
E_a (kJ.mol^{-1})	33.05	42.65
D_0 x10^{-4} (m^2.s^{-1})	1.024	5.332

temperatures and pellets size increase. The activation energy increases with the increasing of pellets size and its values were 33.05 kJ/mol for d_1 and 42.65 kJ/mol for d_2.

Nomenclature: a,b,c,n, Coefficients in models; **k, k_0, k_1, g, h,** Constants in models (s^{-1}); **D_{eff},** effective moisture diffusivity (m^2/s); **D_0,** effective moisture diffusivity for an infinite temperature (m^2/s) **E_a,** activation energy (kJ/mol); **X_e,** equilibrium moisture content (kg water/kg dry matter); **X_0,** initial moisture content (kg water/kg dry matter); **X_R,** moisture ratio; **X_t,** moisture content at T time (kg water/kg dry matter); **$X_{Rexp,i}$,** experimental moisture ratio; **$X_{Rpre,i}$,** predicted moisture ratio; **N,** number of observation; **p,** number of parameters in a model; **r,** radius; **RMSE,** root mean square error; **R,** universal gas constant (J/mol.K); ***R^2*,** coefficient of determination; ***T*,** drying temperature (°K); ***t***, drying time (s).

Greek letter: χ^2, Chi-square.

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