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APPLICATION OF MATHEMATICAL MODELS TO PREDICT THE DRYING KINETICS OF ENERGY CANE

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Abstract. *This study aims to evaluate drying kinetics of the energy cane Saccharum spontaneum* L. *as a pre-treatment of pyrolysis process. Isothermal drying experiments were performed in a forced air oven applying four temperatures: 30 °C, 60 °C, 90 ºC, and 103 °C. The biomass samples were cut in cylinder geometry with 0.01 m of diameter and 0.05 m of length. The drying kinetics curves (moisture ratio versus time) were fitted using semi-theoretical models. The activation energy (E) and the pre-exponential factor (A) were determined using two methods. In the first method, the parameters were determined by Arrhenius and Fick's laws dependent of the effective diffusivity coefficient. In this case, infinite cylinder was assumed obtaining the effective diffusivity between* 2.5×10^{-10} $m^2.s^{-1}$ *and* 5.2×10^{-9} $m^2.s^{-1}$ *for the all temperature: 39.8* $kJ.mol^{-1}$ *and* 2.14×10^{-3} $m^2.s^{-1}$ *for E and A, respectively. In the second method, the parameter k of each semi-theoretical model was linearized as a function of temperature according to Arrhenius law obtaining the E between 41-42.5* $kJ.mol^{-1}$ *and the A between 6.09* s^{-1} *and 6.32* s^{-1}*. The activation energies showed a reliable correlation (6.5 %) between the methods studied.*

Keywords: *Arrhenius law, biomass, effective diffusivity, Fick´s second law, semi-theoretical model*

1. INTRODUCTION

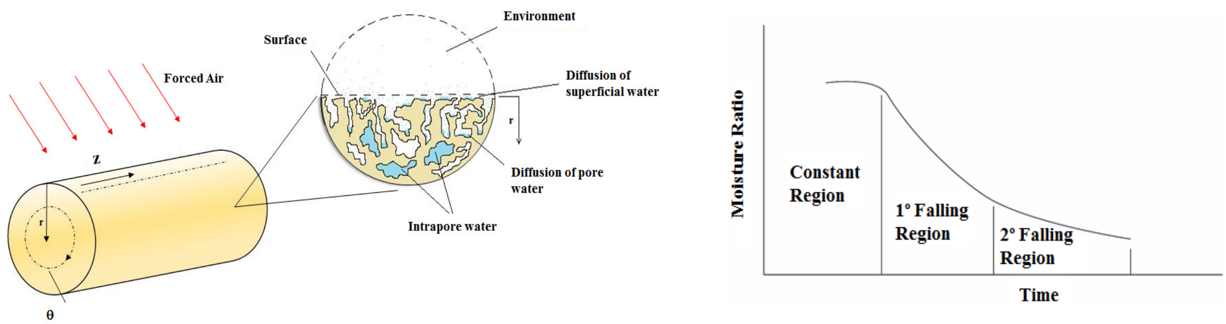
The sugarcane is being developed and improved in several programs around the world targeting on the increase of sugar yields per unit area, on the other hand nowadays this raw material is considered an excellent feedstock for bioenergy. There are two main paths to improve the sugarcane energy performance from the point of view of useful energy per unit of cultivated area: First, increasing the conversion efficiency of primary energy to useful energy forms (better distillery conversion of sugars in ethanol and better use of the fibers). Second, develop sugarcane varieties for energy production (Leal *et al*., 2013). Energy Cane is a biomass with a higher fiber content compared to traditional sugarcane. In recent years, this biomass has been used as raw material for second generation ethanol production and thermal conversion processes (TCP). In TCP's is necessary a pre-treatment (drying) in which the removal of moisture is performed in order to increase the heating value of the biomass. The study and knowledge of drying kinetic parameters is relevant for the equipment designs and optimization of industrial operations. Then, the purpose of this work is to study the drying kinetics of the Energy Cane *Saccharum spontaneum* Linnaeus as a pre-treatment, aiming to perform the pyrolysis processes. Furthermore, the kinetic parameters applying semi-theoretical models and the Fick's second law were compared.

2. LITERATURE REVIEW

2.1 Diffusion theory

The diffusion phenomena of drying of energy cane represented by cylindrical shape is showed in Fig. 1a, including the diffusion of the superficial water, molecular diffusion, diffusion through solid pores and vapor diffusion in air-filled pores. These effects are usually represented by the effective diffusivity (Senadeera *et al.,* 2003).

The drying process of a porous material could be represented experimentally relating the moisture ratio and drying time by three main stages, according to Fig. 1b. The first stage concerns to a constant region in the drying curve denoting the absorption of the latent heat on the solid surface, in which the free moisture of the material is moved by the capillary forces reaching the saturation moisture. In the next step, the surface drying of the sample can be observed in the first falling period (Treybal, 1988). In the second range of the falling rate period, the diffusion of water decreases and occurs the increase of the internal resistance of the water removal until solid reaches the equilibrium moisture (Jankowsky and Santos, 2004).



(a) Scheme of water diffusion (b) Characteristic drying curve

Figure 1. Drying processing of porous material

2.2 Drying kinetics modeling

The moisture and vapor migration during the drying process is controlled by the diffusion (Senadeera, *et al.,* 2003), and can be represented by the Fick's second law in Eq. (1),

$$\frac{\partial m}{\partial t} = D_{eff} \nabla^2 m \quad (1)$$

where, D_{eff} is the effective diffusivity, (m^2/s); t the time, (s), and m the sample mass, (g).

The Eq (1) can be written by Eq. (2) considering cylindrical coordinates:

$$\frac{\partial m}{\partial t} = -D_{eff}\left(\frac{\partial^2 m}{\partial {r_p}^2} + \frac{1}{r_p}\frac{\partial m}{\partial r_p} + \frac{\partial^2 m}{\partial z^2} + \frac{1}{{r_p}^2}\frac{\partial^2 m}{\partial \theta^2}\right) \quad (2)$$

where, r_p is the particle radius, (m); z, the length of the particle, (m); and θ, the angular coordinate (-)

The Fick's second law has simple analytical solutions considering uniform initial moisture, negligible shrinkage, external movement resistance, and heat transfer effect. Also, the variation of water mass in the θ and z directions, and initial thermal transient effect are negligible. Applying all considerations, the Eq. (3) is found, where MR represents the moisture ratio defined in terms of the moisture content (m) at time t, the initial moisture content (m_o), and the equilibrium moisture content (m_e).:

$$MR = \frac{m - m_e}{m_o - m_e} = \sum_{n=1}^{\infty} \frac{4}{\beta_n^2} exp\left[-\frac{\beta_n^2 D_{eff} t}{r_p^2}\right] \quad (3)$$

The right side of the Eq. (3) represents the solution of the second Fick's Law in terms of Bessel's function being β_n, the Bessel's roots. Applying the first term of the series of Bessel's function, the Eq. (4) represents the analytical solution of Eq. (3), in which the moisture ratio is dependent of the effective diffusivity.

$$MR = \frac{4}{\beta^2} exp\left[\frac{-\beta^2 D_{eff}}{r_p^2} t\right] \quad (4)$$

Although the classic theoretical mass transfer phenomena (Fick's Law) can be applied to describe drying kinetics, semi-theoretical models were established in order to simplify the differential equation's solution series (Gomez de la Cruz *et al.,* 2014).Table 1 shows the models with one term (Lewis 1921, Page 1949, Henderson and Pabis 1961, Logarithmic 2005) and with two terms (Two-term exponential 1979, Two-term 1986, Approach of diffusion 2001) considering a thin-layer drying of particles.

Table 1. Drying kinetics semi-theoretical models (*adapted of Souza, 2015*).

Semi-theoretical models		Equation	Constants	Author
1	Lewis	$MR = exp(-kt)$	k	Lewis (1921)
2	Page	$MR = exp(-kt^n)$	k, n	Page (1949)
3	Henderson and Pabis	$MR = a\,exp(-kt)$	a,k	Henderson and Pabis (1961)
4	Two-term exponential	$MR = a\,exp(-kt) + (1-a)exp(-kat)$	a, k	Sharaf-Eldeen (1979)
5	Two-term	$MR = a\,exp(-k_0 t) + c\,exp(-k_b t)$	a, c, k_0, k_b	Noomhorn & Verma (1986)
6	Approach of diffusion	$MR = a\,exp(-kt) + (1-a)exp(-kbt)$	a, k, b	Yildiz *et* al (2001)
7	Logarithmic	$MR = a\,exp(-kt) + c$	a, k, c	Akgun & Doymaz (2005)

2.3 Drying kinetics applications

The diffusion in the drying process of food residues and biomass is mainly represented by semi-theoretical models, adjusting constants (a, b, c, k, k_0, k_b, n) observed in Tab. 1, with the experimental data. However, the determination of drying kinetic parameters (activation energy, pre-exponential factor) can be estimated by two paths: the second Fick's law related to effective diffusivity coefficient (theoretical model) or the semi-theoretical models and the Arrhenius law.

Senadeera *et al.* (2003) studied the drying kinetics of vegetable residues (potatoes, beans, peas) using three different geometries (cylinder, parallelepiped, and sphere, respectively) for three temperatures (30°C, 40°C, 50°C) using a fluidized bed dryer. The activation energy of each residue was 14.34 kJ.mol^{-1}, 39.41 kJ.mol^{-1} and 42.35 kJ.mol^{-1}, respectively applying the second Fick's Law. Concerning the application of the semi-theoretical models of Lewis (1921) and Page (1949) for obtaining drying constants, and the Page model showed be more accurate with experimental data. In this case, the activation energies were 24.27 kJ.mol^{-1} for potatoes, 29.57kJ.mol^{-1} for beans and 58.15 kJ.mol^{-1} for peas.

Akgun e Doymaz (2005) studied the drying kinetics for olive residues (composed by the pulp and stone) using a thin layer of 0.8 cm thickness (250 ± 0.2g), under the drying temperatures between 50°C and 110°C in a cabinet dryer. Comparative moisture ratio versus time curves were elaborated applying different semi-theoretical models such as: models 1, 2, 3, 6, 7 showed in the Tab. 1, Page with modifications (1973), and Wang and Singh (1978). The authors concluded that the Logarithmic model represents better the drying of this residue. The activation energy, 17.97 kJ.mol^{-1}, was determined using the effective diffusivity and second Fick's Law.

Costa (2008) studied the drying kinetics of sugarcane bagasse (particles retained in the sieve 4 Mesh) using a tray coupled in a fixed bed, with 3.5 cm of layer of particle biomass applying 50°C, 60°C and 70°C of temperature. The same semi-theoretical models discussed by Akgun e Doymaz (2005), with replacement of model 6 by model 5, were used to represent the diffusion phenomena being the Page's model the most accurate. The semi-theoretical model constants were presented, however the activation energies were not defined.

3. MATERIALS AND METHODS

3.1 Materials

The age of the energy cane (specie *Saccharum spontaneum* Linnaeus) applied in this study was 18 months. This cane was collected manually, removing the straw and the superior stalk, to avoid high content of phenols. The stalk was cut in cylinder geometries (1cm of diameter and 5cm of length) with initial moisture content of 48.8 ± 0.96 % w/w (*wet basis*). Physical, chemical and thermal properties were determined using 246 ± 6.30 µm of particle diameter as reference: residual moisture content (4.73 ± 0.63 % w/w, *wet basis*) obtained through the thermogravimetric analyzer; true density (1424.3 kg.m^{-3} ± 1.53) by helium pycnometry method; the apparent density (785.0 kg.m^{-3}, d_p=1cm) by mercury porosimetry; the higher heating value (17.31 MJ.kg^{-1} ± 0.07) and the lower heating value (15.57 MJ.kg^{-1}) applying the ASTM D-240 and Mendeleev equation (1949), respectively; the ultimate analysis was 43.53 % ± 0.16 of carbon, 6.33 % ± 0.0 of hydrogen, 0.08 % ± 0.0 of nitrogen, and 52.42 % of oxygen by elemental analyzer; the proximate analysis with values of 85.00 % ± 1.3 of volatile material content (ASTM E 1755), 1.17 % ± 0.49 of ash content (ASTM E 872), and for by difference 13.83 % ± 0.93 of fixed carbon content.

3.2 Experimental procedure

Isothermal drying experiments were carried out in a forced air oven Q314M242 (Quimis, Diadema, São Paulo, Brazil) applying 30°C, 60°C, 90°C and 103°C of temperature. Temperatures above 103°C were not used to avoid the volatilization of the material.

The drying oven was turned on, one hour before starting the experiments to attain the thermal equilibrium inside the equipment. Firstly, the weight of the aluminum plates (3.5-4 g e 12.5 cm of diameter) was measured and after that 7 g to 10 g of fresh biomass were added. Drying tests were performed in duplicate. The samples were exposed to drying conditions for 5 min followed by 5 min of stabilization in a glass desiccator to reach ambient temperature. The weight of the sample was recorded in a weighting scale MARK210 (BEL Engineering, UK). This procedure was repeated until the weight reached the equilibrium. The sample masses collected for each time represents the drying kinetics curves, being defined by the moisture ratio showed in the left side of Eq. (3) and validated by the semi-theoretical models (Tab. 1).

$$MR_{exp} = \frac{m - m_e}{m_o - m_e} \tag{3}$$

3.3 Determination of drying kinetics parameters, *E*, *D*$_0$, and A

3.3.1 Analysis of the Fick´s second law

Using the moisture ratio curves obtained experimentally (see experimental procedure), the activation energy could be calculated theoretically with the Fick's second law. For this purpose, it was necessary to estimate the effective diffusivity of the material for each temperature performing a linearization of the drying curves through moisture ratio as function of time (Eq. 4). With the values obtained in this linearization, the Bessel's root (β) was obtained by the intercept and the effective diffusivity with the slope of the linear equation, as is showed in Eq. (4).

$$Ln(MR) = \frac{-\beta^2 D_{eff}}{r_p^2} t + Ln\left(\frac{4}{\beta^2}\right) \tag{4}$$

The effective diffusivity is written as Arrhenius type equation, Eq. (5), which depends of the diffusivity coefficient (D_0) at infinite temperature in ideal condition, activation energy (E), ideal gas constant (R) and the absolute temperature (T),

$$D_{eff} = D_0 \exp\left(\frac{-E}{RT}\right) \tag{5}$$

Then, the activation energy was obtained by the slope of linearization function, Eq. (5), and the result is showed in Eq. (6).

$$Ln(D_{eff}) = \frac{-E}{RT} + LnD_0 \tag{6}$$

The activation energy showed in the Eq. (6) represents the energy that the particles must attain in order to trigger the drying process.

3.3.2 Analysis of the semi-theoretical models

Initially, the constants of the semi-theoretical models showed in Tab. (1) were obtained applying generalized reduced gradient (GRG) algorithm for optimizing nonlinear adjustment. In order to run the iterative calculation of the GRG method, the models were compared with experimental data of moisture ratio using the least squares method (Eq. 7) minimizing the difference by means of Microsoft Excel's 2007 add-in Solver.

$$\sum (MR_{exp} - MR_{theo})^2 = 0 \tag{7}$$

where, MR_{exp} is the experimental value of moisture ratio MR, [-]; MR_{theo}, the theoretical value of moisture ratio MR, calculated for each semi-theoretical model [-].

Once the constants were adjusted, the drying velocity (k) of Lewis (1921), Logarithmic (2005) and Henderson & Pabis (1961) models can be written as the Arrhenius law, according to the Eq. (8).

$$k(T) = A\,exp\left(\frac{-E}{RT}\right) \tag{8}$$

The drying kinetic parameters were also calculated by the linearization function of the Eq. (8), plotting the drying velocity (k) as a function of the inverse of absolute temperature, as shown in Eq. (9),

$$Lnk(T) = \frac{-E}{R}\left(\frac{1}{T}\right) + LnA \tag{9}$$

The models Lewis (1921), Logarithmic (2005) and, Henderson and Pabis (1961) were applied to compare the activation energy calculated with the Fick's second law. In order to determinate the deviation between theoretical data from semi-theoretical models and experimental data, the correlation coefficient (r) and the root-mean-square deviation (RMSE) were applied.

4. RESULTS AND DISCUSSION

4.1 Analysis of drying curves

Figure 2 shows the evolution of moisture ratio as a function of time for each temperature from 30 ºC to 103 ºC. As can be seen, the higher the temperature, the lower the total drying time. At the beginning of the drying process, a higher rate of moisture loss was observed. Subsequently, this rate decreases substantially, because the internal resistance increases, leading two thirds of the total drying time to remove 30 % of moisture content remained in the particle.

4.2 Modelling of drying curves

In this study, the semi-theoretical models (Tab. 1) were applied to represent the diffusion phenomena in the falling rate region (see Fig. 1) considered in the whole drying time, and represented in Fig. 2. The Fig. 2 (a), (c), (e), (g) show the one term models and Fig. 2 (b), (d), (f), (h) show the two term models for 30 °C, 60 °C, 90 °C and 103 °C . Most of the models analyzed overestimate the late stage of drying, mainly in the temperatures of 30°C and 60°C. To verify the accuracy of the approach done by the semi-theoretical models, the determination coefficient (r), and root mean square error ($RMSE$), were calculated, obtaining values between 0.995-0.999, and 2.53 10^{-5}-1.22 10^{-3}, respectively.

Since the semi-theoretical models have two terms of the series solution (analytical) of Fick's second law, they provide higher accuracy representing physically the drying process. The diffusion model approach was the best adjustment to represent the experimental data, obtaining 0.999 of determination coefficient (model 6).

4.2.1 Determination of kinetic parameters (E, A) using the Fick´s second law

The effective diffusion coefficient points out the difficulty that water has to be diffused through the material. This coefficient can be used to estimate the time necessary to obtain certain moisture content (Gomez de la Cruz *et al.,* 2014). The effective diffusivity was obtained through Eq. (7) and the results are presented in Tab 2, which the values were between 2.56 10^{-10} and 5.18 10^{-9} $m^2.s^{-1}$.

Table 2. Activation energy and pre-exponential factor calculated with second Fick's Law from literature and comparative with this work

Author	Material	T (°C)	D_{eff} ($m^2.s^{-1}$)	D_0($m^2.s^{-1}$)	E (kJ.mol^{-1})
Present work	Energy Cane	30	2.56 10^{-10}	2.14 10^{-3}	39.89
		60	1.41 10^{-9}		
		90	4.36 10^{-9}		
		103	5.18 10^{-9}		
Madamba *et al*. (1996)	Garlic	50-90	2.02 10^{-10}- 4.24 10^{-10}	20.2 10^{-11}	54.9
Lopez *et al*. (2000)	Vegetables waste	50-150	6.03 10^{-8}- 3.15 10^{-8}	9.82 10^{-6}	19.82
Doymaz and Pala (2003)	Corn	55-75	9.488 10^{-11} - 1.768 10^{-10}	6.14 10^{-6}	29.56
Akgun *et al*. (2005)	Olive cake	50-110	3.382 10^{-9}- 1.134 10^{-8}	3.128 x10^{-6}	17.97

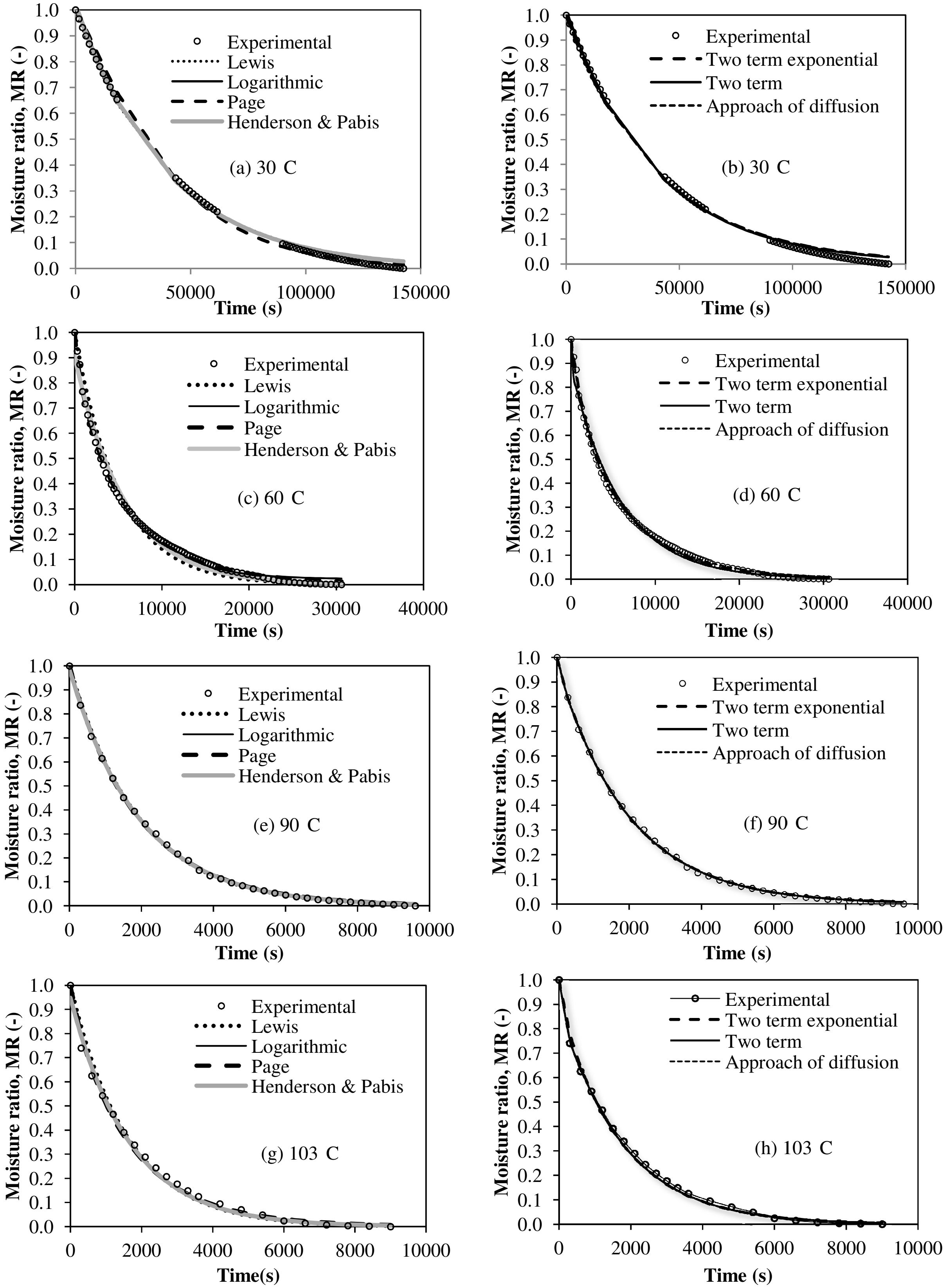


Figure 2. Moisture ratio as a function of time – comparasion between semi-theoretical and experimental data for energy cane - One term models (a, c, e, g), Two term models (b, d, f, h)

Using the Arrhenius type equation to describe the dependence of effective diffusivity on the temperature, values of 39.89kJ/mol for activation energy and 2.14 x10^{-3} $m^2.s^{-1}$ for pre-exponential factor were found.

Due to the diversity of characteristics (initial moisture content, porosity, etc) of the materials presented in Tab. 2, the activation energy is different in each case. The activation energy for *Saccharum spontanuem* L. is lower than the energy required for garlic drying, but higher than vegetable waste, corn and olive cake.

4.2.2 Determination of kinetic parameters (E, A) using the semi-theoretical models

In the Tab.3 is presented the drying velocity constant, pre-exponential factor and the activation energy for the different temperature (30 °C-103 °C) calculated by Lewis (1921), Henderson and Pabis (1961), and Logarithmic (2005) models.

The model that performed a better linearization adjustment (Eq. 9), r= 0,975, was the Henderson and Pabis model, which shows an activation energy of 41.73 $kJ.mol^{-1}$ and a pre-exponential factor of 6.15 s^{-1}. However, the average activation energy was quite similar of 42 $kJ.mol^{-1}$, because the drying velocity constant obtained with each model presented.

Comparing this value with the kinetic parameters obtained using Fick's second law (effective diffusivity), the difference was below 6.5 %, showing a good agreement between the models applied.

Besides that, our results can corroborate with Costa (2008) work found to sugarcane bagasse (44.78$kJ.mol^{-1}$) applying the Logarithmic model.

Table 3. Values of activation energy and pre-exponential factor calculated with semi-theoretical models

Semi-theoretical Model	T (°C)	k (s^{-1})	A (s^{-1})	E ($kJ.mol^{-1}$)
Henderson & Pabis	30	2.54 10^{-5}	6.13	41.73
	60	1.75 10^{-4}		
	90	5.13 10^{-4}		
	103	6.00 10^{-4}		
Logarithmic	30	2.54 10^{-5}	6.09	41.52
	60	1.91 10^{-4}		
	90	5.13 10^{-4}		
	103	6.00 10^{-4}		
Lewis	30	1.00 10^{-4}	6.32	42.14
	60	2.49 10^{-4}		
	90	6.00 10^{-4}		
	103	9.00 10^{-4}		

5. CONCLUSION

This paper aimed to analyze the drying process and the kinetic parameters of the energy cane (*Saccharum spontaneum* L.), comparing with two different kind of models. Concerning to the drying process, seven semi-theoretical models were performed and presented an accurate approach (r = 0.995-0.999). Nevertheless, the semi-theoretical models with two terms better represent the diffusion phenomena in the drying falling rate of biomass particles. For the case of kinetic parameters, the activation energy and the pre-exponential factor were 39.89 $kJ.mol^{-1}$ and 2.14 x$10^{-3}m^2.s^{-1}$ respectively calculated with the Fick's second law, and by semi-theoretical models were 42 $kJ.mol^{-1}$ and 6.21 s^{-1}, respectively. Finally, the activation energies obtained with the two methods are comparable (< 6.5 %), and it can be applied to represent the diffusion phenomena of drying process.

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