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# **Research Paper**

# Arrhenius activation energy for water diffusion during drying of tomato leathers: The concept of characteristic product temperature



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Keywords: Tomato leather Activation energy Drying Mathematical modelling The drying kinetics of tomato leathers was studied between 40 and 80 °C and the corresponding thermal histories were recorded. As air and product temperatures were found to be considerably different during the drying runs, the original concept of "characteristic product temperature" was proposed. It is defined as the temperature corresponding to the time at which the instantaneous drying rate is equal to the mean drying rate. Then, the Arrhenius activation energies  $(E_a)$  obtained by two methods were compared: (1) correlating the diffusion coefficient (D) with the air temperature and (2) correlating D with the characteristic product temperature. Drying curves comprised a "wet zone", where moisture content fell almost linearly with time, and a subsequent "dry zone". In the first zone, the Arrhenius relationship with the air temperature yielded an  $E_a$  of 18.6 kJ mol<sup>-1</sup> whereas, when utilising the characteristic product temperature the activation energy was 47.3 kJ mol $^{-1}$ , comparable to the enthalpy of water desorption. In the second zone, activation energies were 21.7 and 26.6 kJ mol<sup>-1</sup>, respectively. Molecular mobility theory suggests a stronger temperature dependence of diffusion-limited phenomena in high moisture content matrices and thus would forecast a higher  $E_a$  for the wet zone. Therefore, the characteristic product temperature is more meaningful than the air temperature in the Arrhenius relationship.

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# 1. Introduction

Drying kinetics studies provide useful information about product drying for industry, contributing also to the knowledge of the drying mechanisms as well as to more practical issues such as dryer design (Ratti, 2001). The application of physically-based models to experimental data of drying kinetics provides mass transfer parameters such as water diffusion coefficients in the product matrix, which can

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Nomenclature	t <sub>c</sub> characteristic time (min)
$a_w$ water activity $Bi_m$ mass transfer Biot numberDwater diffusion coefficient (m² s <sup>-1</sup> )daverage product thickness (m) $E_a$ activation energy (kJ mol <sup>-1</sup> ) $h_r$ relative humidity of the drying air (%) $m_i$ initial product mass (kg) $m_t$ product mass at time t (kg) $r^2$ coefficient of determination $T_a$ air temperature (°C) $T_c$ characteristic product temperature (°C) $T_c$ absabsolute characteristic product temperature (°K)ttime (min)	tlength of time of the wet zone (min) $W_{ad}$ dimensionless moisture content $W_e$ equilibrium moisture content (kg [water] kg^{-1} [dry matter]) $W_f$ final moisture content (kg [water] kg^{-1} [dry matter]) $W_i$ initial moisture content (kg [water] kg^{-1} [dry matter]) $W_t$ moisture content at time t (kg [water] kg^{-1} [dry matter]) $W_t$ moisture content at time t (kg [water] kg^{-1} [dry matter]) $W_t$ moisture content at time t (kg [water] kg^{-1} [dry matter]) $W_t$ moisture content at time t (kg [water] kg^{-1} [dry matter]) $\beta_n$ roots of the equation $\beta_n \cdot tg \cdot \beta_n - Bi_m = 0$

be utilised in drier simulation models to predict the drying time and, hence, equipment dimensions (Crapiste & Rotstein, 1997).

Dehydration of fresh foods is a complex process that involves simultaneous phenomena of heat and mass transfer in the product. Moisture migration in the food may be governed by a combination of mechanisms, though at moderate drying temperatures, liquid diffusion is dominant. Pezzutti and Crapiste (1997) carried out an experimental and theoretical study of garlic drying. Their mathematical modelling was based on a Biot-dependent analytical solution of the unsteady state differential diffusion equation, i.e., they modelled a situation that is not totally controlled by either internal or external resistances to mass transfer. The authors used only one term of the solution mentioned above, so that it can be plotted as a straight line in a semi-log graph. On the other hand, most food drying researchers have correlated their diffusion coefficients with the reciprocal of the absolute air temperature, which is normally kept constant during kinetic studies (Zogzas, Maroulis, & Marinos-Kouris, 1996). This method may be satisfactory in low moisture products, such as cereals or oilseeds, where the product temperature rapidly approaches thermal equilibrium with the air temperature. However, in high moisture foods, as fruits, vegetables or formulations based on these, product temperature stays well below air temperature for a considerable fraction of the drying time. Therefore, in order to evaluate drying kinetics of highmoisture foods based on analytical solutions of the mass transfer phenomenon, the proposed hypothesis is that, if the activation energy is expected to have some physical meaning, the relationship between the diffusion coefficient and temperature must be established with a representative value of product temperature instead of with the air temperature. In this work, we have proposed the original concept of "characteristic product temperature", defined as that corresponding to the time at which instantaneous drying rate is equal to the mean drying rate. This aspect may have more practical consequences than it would appear at first sight: as numerically or analytically-solved thin layer drying models may be utilised in dryer simulation computer programs that also include variations of air temperature and humidity, the utilisation of accurate values of the activation energy may lead to improved

predictions of the effect of air temperature on the energy consumption for drying and thus of fuel requirements of natural gas, biodiesel, LPG, etc., for a given period of time.

The objective of this work was, therefore, to compare the Arrhenius activation energy  $(E_a)$ , as determined by two methods: (a) correlation of the effective water diffusion coefficient with the reciprocal of the absolute air temperature and (b) correlation with the reciprocal of the absolute characteristic product temperature. This comparison was carried out at high and low moisture contents during drying of tomato leather, a snack product which has potential for use in different pizza variations, as well as in sandwiches and a variety of snacks. A previous paper on drying of tomato leather dealt with moisture curves alone in order to find a suitable mathematical model (Fiorentini, Leiva Diaz, & Giner, 2008). In the present work, simultaneous thermal and moisture histories were newly measured in order to develop diffusion coefficient relationships with air as well as with product temperatures.

# 2. Materials and methods

### 2.1. Sample preparation

Pear-shaped, Italian type tomato was used to prepare tomato purée. The tomato leather formulation was designed so that the saccharide-acid-high methoxyl pectin gelation can occur during drying as indicated in previous research (Fiorentini et al., 2008). The initial material for studying drying kinetics presented a fluid-like consistency, with the following approximate composition as calculated by mass balance (% w/ w): moisture content, 88.20; polydextrose, 5.75; sugars, 2.60; protein, 1.10; added pectin, 1.00; fibre, 0.90; citric acid, 0.25; and minerals, 0.20.

# 2.2. Initial moisture content of the formulation

The moisture content of the formulation was determined by the AOAC method 984.25 (AOAC, 1998) at atmospheric pressure and 105  $^{\circ}$ C up to reaching constant weight in a Mettler LP 16 moisture analyser with a readability of 0.01 g. Eight replicates were measured. A 10 g sample was placed in the analyser and the expected error was then  $100 \times 0.01$  g/8.8 g, i.e. around 0.11%. Accepted methods for oven moisture determinations tolerate errors of up to 0.3% (Greensmith, 1998) so that the technique used in this work was well below the limit.

# 2.3. Drying experimental technique

The formulation was placed in square stainless steel trays 0.20 m side and 0.02 m deep, with an initial product thickness of  $7.5 \times 10^{-3}$  m. Trays were placed inside a pilot scale, purposedeveloped dryer, with an in-situ weighing system (Fig. 1). The drying chamber contained a framework holding up to three trays. To avoid weighing instabilities caused by the airflow, a by-pass valve diverted the air current to a parallel duct running above the dryer while the framework was disengaged by a magnetic system and deposited onto the digital scale. Once the mass was read, the flow was restored and the framework lifted and engaged. Drying runs were conducted in duplicate, at air temperatures of 40, 60 and 80 °C, with a controlled air velocity of 2.0  $\pm$  0.1 m s<sup>-1</sup>, as usually found in industrial dryers (Greensmith, 1998). Knowing the initial moisture content (W<sub>i</sub>), which was measured as indicated above, subsequent values during drying were calculated by mass balance, assuming constant dry matter in the product throughout the process (Eq. (1)).

$$W_t = \frac{m_t(1+W_i)}{m_i} - 1$$
 (1)

 $W_t$  and  $m_t$  are the moisture content (kg [water] kg<sup>-1</sup> [dry matter]) and the product mass (kg), respectively at time t, while  $W_i$  and  $m_i$  are the corresponding initial values. Experimental data for the product mass were obtained from the weighing system at several times, using an OHAUS digital scale (readability, 0.01 g; weighing capacity, 3000 g, OHAUS Corporation, USA).

#### 2.4. Operating conditions for the drying runs

Drying experiments were conducted in a pilot plant. Room temperature and relative humidity were measured simultaneously during the drying experiments using a Testo 608H2 thermohygrometer with digital display (Testo AG, Germany). Results were averaged and supplied to the Akton psychrometric software (Akton Associates Inc., USA) to determine the relative humidity ( $h_r$ ) of the drying air. The equilibrium moisture content at each drying condition ( $W_e$ ) was calculated from the Halsey model (Eq. (2)) (Chen & Mujumdar, 2008, 2006), previously fitted for the product isotherm at 40 °C with a coefficient of determination of 0.999 (Demarchi, Quintero Ruiz, De Michelis, Giner, 2013):

$$W_{e} = \left(\frac{-0.083}{\ln(a_{w})}\right)^{\frac{1}{0.990}}$$
(2)

where  $a_w$ , the water activity of the product, is assumed to be equal to  $h_r/100$  at equilibrium conditions. In general, isotherms of fresh fruits and leathers are not affected by temperature at water activities below 0.15 (Demarchi, Quintero Ruiz, De Michelis, & Giner, 2013; Kaya & Kahyaoglu, 2005; Mrad, Bonazzi, Boudhrioua, Kechaou, & Courtois, 2012), such as those used to calculate the moisture content in equilibrium with the drying air conditions ( $W_e$ ). This consideration allows the calculation of  $W_e$  by Eq. (2) for all drying temperatures, as the relative humidity of the drying air ( $h_r$ ) never exceeded 13.2%, and so the product  $a_w$  at equilibrium was never higher than 0.132 at the drying experimental conditions.

# 2.5. Measurement of product surface temperature during drying

Product surface temperature curves were measured by a noncontact infrared thermometer Testo 830 T2. At several times during drying runs, the instrument was pointed at the product surface through an orifice in the dryer roof, located above the



Fig. 1 – Diagram of the pilot scale dryer. A: resistance heaters; B: air deflector; C: air bypass; D: digital temperature and velocity control; E: digital scale for in situ weighing; F: drying cabinet; G: air temperature sensors; H: orifice for non-contact measurement of product temperature.

sample. The orifice was normally closed with a polypropylene plug, which was removed for a few seconds to allow the measurement. Each temperature was an average of five determinations recorded over 10 s. Application of such a technique was conducted on counter-flow drying of wheat by Bruce and Giner (1993).

# 3. Modelling and calculations

# 3.1. Analysis of the kinetic behaviour

Drying curves were modelled by considering two different zones, according to their kinetic behaviour: the first period, named here the "wet zone", corresponds to an apparently constant drying rate while the second part of the curve, the "dry zone", is represented by a falling drying rate.

To determine the length of the wet zone period (t), coefficients of determination  $(r^2)$  for linear regression after the two first points of each drying curve were observed, with increasing number of data points, as experimental values of  $W_t$  were included one by one. A point was considered to be the end of the wet zone when the inclusion of the following point to the model resulted in a decrease of 0.0018 or more units in  $r^2$ .

# 3.2. Modelling of the drying curve

For each zone of the drying curve, the analytical solution of the unsteady state diffusion equation of water in a solid (Crank, 1975), integrated over the leather thickness, was employed (Eq. (3)), which predicts the mean product moisture content ( $W_t$ ) as a function of time (t) for the general situation where both internal and external resistances to mass transfer are relevant.

$$W_{ad} = \frac{W_t - W_e}{W_i - W_e} = \sum_{n=1}^{\infty} \frac{2Bi_m^2 \exp\left[\frac{-\beta_n^2 Dt}{d^2}\right]}{\left(\beta_n^2 + Bi_m^2 + Bi_m\right)\beta_n^2}$$
(3)

The mass transfer Biot number ( $Bi_m$ ), which is a dimensionless parameter expressing the ratio of the internal to the external resistance to the mass transfer rate, and the water diffusion coefficient (*D*) are the fitting parameters. In turn, *d* is the average leather thickness for each drying zone, while  $\beta_n$  stands for the *n*th root of the following transcendental equation (Eq. (4))

$$f(\beta_n) = \beta_n \operatorname{tg}(\beta_n) - \operatorname{Bi}_m = 0 \tag{4}$$

A correlation between the first six roots  $\beta_n$  and the mass transfer Biot number has been developed in previous work (Leiva Díaz, Giannuzzi, & Giner, 2009) and is utilised here. For a formal definition of the mass transfer Biot number, the reader is referred to Crapiste and Rotstein (1997). The solution appearing in Eqs. (3) and (4) has rarely been employed in the food research literature, despite its solid foundation on the diffusion mechanism. By using the named correlation, Eqs. (3) and (4) were programmed in the MATLAB environment (MATLAB, 2007) and it was shown that the first two terms of the series provided, in terms of  $r^2$ , enough accuracy for both zones of the curve, so Eq. (5) was utilised to model each drying zone.

$$W_{ad} = \frac{W_{t} - W_{e}}{W_{i} - W_{e}} = 2 \left[ \frac{Bi_{m}^{2} \exp\left[\frac{-\beta_{1}^{2}Dt}{d^{2}}\right]}{\left(\beta_{1}^{2} + Bi_{m}^{2} + Bi_{m}\right)\beta_{1}^{2}} + \frac{Bi_{m}^{2} \exp\left[\frac{-\beta_{2}^{2}Dt}{d^{2}}\right]}{\left(\beta_{2}^{2} + Bi_{m}^{2} + Bi_{m}\right)\beta_{2}^{2}} \right]$$
(5)

The length of the wet zone corresponds to the final time of the wet zone and to the initial time of the dry zone.

A statistical program (Systat, 1990) was utilised to fit the model to the experimental data using nonlinear regression, obtaining the values of  $Bi_m$  and D corresponding to the wet and dry zones of each drying curve, measured at constant air temperature.

# 3.3. Calculation of the characteristic product temperature

As air and product temperatures were found to be considerably different during the drying runs, it was necessary to devise a method to calculate a representative value of the product temperature, for both the wet and dry zones of the curve. We propose the original concept of "characteristic product temperature" ( $T_c$ ), defined as that corresponding to the time at which the instantaneous drying rate ( $dW_t/dt$ ) is equal to the mean drying rate of the whole zone. In order to calculate that characteristic time ( $t_c$ ), Eq. (5) was rearranged and differentiated to find  $dW_t/dt$  for wet and dry zones (Eq. (6)).

$$\frac{dW_t}{dt} = -2Bi_m^2(W_i - W_e) \left[ \sum_{n=1}^2 \frac{\beta_n^2 D}{\left(\beta_n^2 + Bi_m^2 + Bi_m\right)\beta_n^2 d^2} \exp\left(\frac{-\beta_n^2 Dt}{d^2}\right) \right]$$
(6)

On the other hand, the mean drying rate was calculated from experimental data by Eq. (7) for the wet zone and Eq. (8) for the dry zone.

$$\frac{\Delta W}{\Delta t} = \frac{(W_{t^*} - W_i)}{t^*} \tag{7}$$

$$\frac{\Delta W}{\Delta t} = \frac{(W_f - W_{t^*})}{(t_f - t^*)}$$
(8)

where  $W_f$  and  $t_f$  stand for the moisture content and time at the end of each drying run. The function *fzero* from the MATLAB environment (MATLAB, 2007) was then applied to determine the characteristic time ( $t_c$ ) at which  $dW_t/dt$  takes a value equal to the mean drying rate calculated by Eqs. (7) and (8). Once the  $t_c$  was obtained for each drying zone, the experimental thermal histories were modelled by a *shape-preserving interpolant* (MATLAB, 2007) to find the temperature corresponding to  $t_c$ , i.e., the characteristic product temperature ( $T_c$ ).

#### 3.4. Determination of the activation energy for drying

The values of ln *D* were linearly correlated with the reciprocal of the absolute air temperature  $(T_{a\ abs}^{-1})$  and also with the reciprocal of the absolute characteristic product temperature  $(T_{c\ abs}^{-1})$ , by applying the Arrhenius expression (Eq. (9)):

$$\ln D = \ln D_0 - E_a R^{-1} T^{-1}_{abs}$$
(9)

where  $\ln D_0$  is the pre-exponential factor and  $R = 8.314 \text{ J K}^{-1} \text{ mol}^{-1}$  is the ideal gas constant. Activation energy was calculated from the fitted slope, which corresponds to  $E_a R^{-1}$ .

# 4. Results and discussion

# 4.1. Drying curves and surface thermal histories

As observed in Fig. 2, the drying of tomato leather consisted of an apparently constant rate period, the wet zone, followed by a falling rate period, the dry zone. Likewise, the thermal histories in Fig. 3 showed two distinct asymptotic behaviours: the first, from the initial temperature up to an intermediate value, and the second, which starts as the curve slope increases again, up to another asymptotic value, this time towards the drying air temperature. In general, the product surface tends to reach the air temperature only in the final minutes of the drying process.

The end of the wet zone, when kinetic behaviour changes (dotted vertical line in Fig. 2), corresponds to the time at which the slope of the thermal history begins its second increase (dotted vertical line in Fig. 3). For the studied system, these changes occurred at an average dimensionless moisture content ( $W_{ad}$ ) of 0.33 for all drying temperatures (see Fig. 2 and Table 1). As shown in Eq. (3),  $W_{ad}$  is defined as follows:  $W_{ad} = (W_t - W_e)/(W_i - W_e)$  where  $W_i$  is the initial moisture content,  $W_e$  is the value in equilibrium with the air drying conditions, and  $W_t$  is the mean product moisture content at time t, all expressed as kg [water] kg<sup>-1</sup> [dry matter].

Table 1 shows the values of t and the corresponding experimental values of  $W_{ad}$  for each drying temperature. Calculated values of the characteristic product temperature are listed in Table 2 and also indicated in Fig. 3 for each drying zone.

#### 4.2. Fitting parameters for the wet zone

As discussed in Section 3.2, the first two terms of the Crank's series (Eq. (5)) were found to be as accurate as the complete series (Eq. (3)) in predicting the curve of the average moisture content as a function of time in the wet zone. Therefore, this simplified version was used to fit the mass transfer Biot number and the diffusion coefficient for each drying temperature. The average initial moisture content (Wi) for the formulation was 8.219  $\pm$  0.851 kg [water] kg<sup>-1</sup> [dry matter]. While the initial product thickness was 7.5 imes 10<sup>-3</sup> m, the average thickness d for the wet zone, estimated with a shrinkage model developed earlier (Leiva Díaz et al., 2009), was: 5.0  $\times$  10<sup>-3</sup> m at 40 °C, 4.6  $\times$  10<sup>-3</sup> m at 60 °C, and  $4.7 \times 10^{-3}$  m at 80 °C. Experimental values of W<sub>e</sub> are listed in Table 1, while Table 2 shows the parameters determined by fitting Eq. (5) to experimental data, along with the air and characteristic product temperatures. An optimum value of the Biot number of 0.2 was found for all temperatures in the wet zone of the drying curve, corresponding to a small but not



Fig. 2 – Dimensionless moisture content of the product  $(W_{ad})$  as a function of time, measured at 40 °C (A), 60 °C (B) and 80 °C (C). Vertical dotted lines separate the wet zone (left) from the dry zone (right). Symbols represent experimental data, and lines the model predictions for each zone.

negligible internal resistance to mass transfer. The model predictions for each zone are shown in Fig. 2, along with the experimental drying curves. As Rovedo, Suarez, and Viollaz (1998) pointed out, Biot numbers do not always satisfactorily



Fig. 3 – Product thermal histories for drying air temperatures of 40 °C ( $\bigcirc$ ), 60 °C ( $\bigcirc$ ) and 80 °C ( $\square$ ). Vertical dotted lines separate the wet zone (left) from the dry zone (right). Filled symbols stand for the characteristic time and temperature in each drying zone, while the lines represent the shape-preserving interpolant fitting.

explain the controlling resistance for mass transfer in samples with high initial moisture content (such as this tomato leather formulation), because the sorptional equilibrium at the solid–gas interface is not linear. However, the results found in the literature range from 0.2 to 2.0 (Fiorentini et al., 2008; Giner, 2009; Giner, Torrez Irigoyen, Cicuttín, Fiorentini, 2010; Leiva Díaz et al., 2009). Diffusion coefficients were comparable to those reported for apple leathers (Leiva Díaz et al., 2009) and strawberry leathers (Lee & Hsieh, 2008). Coefficients of determination ( $r^2$ ) indicate good agreement between predictions and data.

# 4.3. Fitting parameters for the dry zone

The prediction of moisture content by Eq. (5) was also highly accurate at long times, i.e., when modelling the dry zone (see Fig. 2). The average thickness d was  $2.45 \times 10^{-3}$  m for all temperatures. Table 2 shows the fitting parameters, as well as the characteristic product temperature in this period. A Biot number of 0.97 was determined for all temperatures, indicating a higher contribution of internal control to water transfer in the dry zone compared with the wet zone.

Table 1 — Operating conditions and parameters of the drying runs.							
T <sub>a</sub> (°C)	h <sub>r</sub> (%)	W <sub>e</sub> (kg [water]	ť	$W_{ad}$ (t <sup>*</sup> )			
		kg <sup>-1</sup> [dry matter])	(min)				
$40 \pm 0.5$	13.2 ± 1.4	0.0397 ± 0.0021	330	$0.3201 \pm 0.0310$			
$60 \pm 0.5$	$6.5 \pm 1.8$	0.0293 ± 0.0031	190	$0.3210 \pm 0.0316$			
$80 \pm 0.5$	$2.6\pm0.1$	$0.0218 \pm 0.0001$	120	0.3575 ± 0.0186			
T , draing oir temperature, by relative humidity of the draing oir							

 $T_a$ : drying air temperature;  $h_r$ : relative humidity of the drying air;  $W_e$ : equilibrium moisture content; t<sup>\*</sup>: length of time of the wet zone;  $W_{ad}$  (t<sup>\*</sup>): dimensionless moisture content for t<sup>\*</sup>.

Table 2 – Results of the fitting of Eq. (5) to data of moisture content of tomato leathers vs time. First two columns include air temperature and characteristic product temperature for each zone of the drying curve.

	T <sub>a</sub> (°C)	T <sub>c</sub> (°C)	Bi <sub>m</sub>	$D (m^2 s^{-1})$	r <sup>2</sup>
Wet zone	40 ± 0.5	30.62	0.20	$6.25 \times 10^{-9}$	0.997
	$60 \pm 0.5$	34.90	0.20	$9.09 imes10^{-9}$	0.997
	80 ± 0.5	44.00	0.20	$1.41  imes 10^{-8}$	0.996
Dry zone	$40 \pm 0.5$	32.12	0.97	$8.55 \times 10^{-10}$	0.998
	60 ± 0.5	53.47	0.97	$1.50  imes 10^{-9}$	0.985
	$80 \pm 0.5$	60.19	0.97	$\textbf{2.19}\times\textbf{10}^{-9}$	0.996

 $T_a$ : drying air temperature;  $T_c$ : characteristic product temperature;  $Bi_m$ : mass transfer Biot number; D: water diffusion coefficient;  $r^2$ : coefficient of determination for Eq. (5).

Diffusion coefficients were observed to be, in general, one order of magnitude lower than in the wet zone, in agreement with results published by Pezzutti and Crapiste (1997) for garlic drying.

# 4.4. Activation energies for wet and dry zones

Figure 4A illustrates the Arrhenius-type graph for the wet zone. Concerning the correlation of the diffusion coefficient with the reciprocal of air temperature  $(T_{a \ abs}^{-1})$ , the conventional method, the activation energy was 18.6 kJ mol $^{-1}$ , comparable to values found by the same method in other foods (Di Scala & Crapiste, 2008; Maskan, Kaya, & Maskan, 2002). Meanwhile, for the characteristic product temperature  $(T_{c,abc}^{-1})$ , the same diffusion coefficients are related to a narrower range on the X-axis and the slope becomes greater, leading to an activation energy of 47.3 kJ mol<sup>-1</sup>. As the characteristic product temperature is more meaningful than air temperature as a value with which the diffusion coefficient can be related, the physical meaning of the larger  $E_a$  value would be better founded. For instance, the activation energy determined for the wet zone is comparable to the enthalpy of water vaporisation, which is around 40 kJ mol<sup>-1</sup>. Such similarity allows an analogy to be explored.

Following the same procedure for the dry zone, natural logarithms of *D* were represented as a function of  $T_{a\ abs}^{-1}$  and  $T_{c\ abs}^{-1}$  (Fig. 4B), to find activation energies of 21.7 and 26.7 kJ mol<sup>-1</sup>, respectively. Not only lower diffusion coefficients, but also a weaker dependency of diffusion-controlled processes on temperature was found for lower moisture contents, for which water mobility is more difficult. This is in agreement with the glass transition theory (Roos, 1995).

The effective diffusion coefficient is, in practice, a transport parameter reflecting the complexity of water migration through the solid matrix as well as desorption, for the temperature range covered in this work. Both phenomena are interdependent and affected by the activation energy.

# 5. Conclusions

By establishing the conventional Arrhenius relationship of the diffusion coefficient (D) with air temperature in the drying of



Fig. 4 – Correlation of the diffusion coefficients for the wet zone (A) and the dry zone (B) with air temperature ( $\bigcirc$ ) and characteristic product temperature ( $\bigcirc$ ). The lines represent the corresponding Arrhenius model predictions.

tomato leather, the activation energy in the wet zone was  $18.6 \text{ kJ mol}^{-1}$ , comparable to values found for foodstuffs by other researchers. However, by correlating *D* with the characteristic product temperature, an activation energy of  $47.3 \text{ kJ mol}^{-1}$  was determined. This value is considered to have a discernible physical meaning, since it is comparable to the enthalpy of water desorption.

On the other hand, when correlating the diffusion coefficient for the dry zone with the air temperature, an activation energy of 21.7 kJ mol<sup>-1</sup> was found. The corresponding relationship with the characteristic product temperatures led to an activation energy of 26.7 kJ mol<sup>-1</sup>.

The difference between 47.3 and 26.7 kJ mol<sup>-1</sup> for the activation energies of wet and dry zone is observed only if the relationship of the diffusion coefficient is established with the characteristic product temperature in each zone. This is consistent with molecular mobility theory, which indicates that the diffusion-limited processes have stronger temperature dependence (i.e., higher activation energy) at higher moisture contents.

These congruent results are not observed when the transport parameter was correlated with the air temperature, a nominal value.

Hence, the proposed concept of the characteristic product temperature may be useful, since it represents the product thermal history better than the drying air temperature and, besides, is related to kinetic parameters through the mean drying rate.

These results also have technological implications for dryer simulation aimed at product design, since the predicted effect of air inlet temperature on specific energy consumption and on product residence time depends on the activation energy.

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