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Additional Information

1 **Influence of temperature, air velocity and ultrasound application on**
2 **drying kinetics of grape seeds**

3
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11
12 **ABSTRACT**

13 The objective of this work was to determine the influence of temperature, air velocity
14 and ultrasound application on the drying kinetics of grape seeds. The drying kinetics were
15 determined at 1.0, 1.5, 2.0 and 3.0 m/s and at 40, 50, 60 and 70°C. At 1.0 and 1.5 m/s, the
16 experiments were carried out with and without ultrasound application. To establish the
17 influence of the variables on the drying kinetics, the results were modeled by means of
18 both the Peleg and a diffusion model. The activation energy was determined (Arrhenius'
19 equation). For an air velocity of over 1.5 m/s, it was determined that the external
20 resistance to mass transfer was negligible. No influence of ultrasound application was
21 observed, probably due to the fact that grape seeds are very hard and have a low level of
22 porosity.

23
24 *Keywords:* Grape seeds; Ultrasound; Air velocity; Effective diffusivity; activation
25 energy

26 INTRODUCTION

27 Spain is the European country which has the largest surface area dedicated to grape
28 cultivation with 1.113 million hectares, 97.4% of which are for the wine industry^[1].

29 By weight, around 13% of the grapes processed in the wine industry end up as a by-
30 product after pressing. The by-product is called grape pomace and it consists of skins,
31 seeds and stems^[2]. According to the literature^[3, 4], the seeds are a rich source of
32 polyphenols and oil. The oil from grape seeds is free from cholesterol and is low in
33 saturated fats. It also has linoleic acid, high density lipoproteins, E vitamin and
34 antioxidants. Thus, it is an oil with interesting nutritional properties.

35 The by-product must be stabilised before extracting the compounds of interest and, to
36 this end, drying is the most commonly used process. Drying stabilizes the raw material by
37 reducing its water content and it also decreases the amount of solvent used in subsequent
38 extraction processes^[5]. Convective drying, using air at different temperatures and
39 velocity, is the most commonly used drying system. The initial moisture content of grape
40 seeds is around 0.82 kg water/kg dry matter and usually drops by between 0.02 and 0.07
41 kg water/kg dry matter. Convective air drying is a highly demanding operation. Thus, in
42 order to reduce energy consumption it is necessary to determine the influence of the
43 process conditions on the dehydration kinetics. In addition, process conditions have an
44 influence on the quality of the final product.

45 One important operating condition is temperature. The dehydration rate increases
46 when the temperature rises due to the fact, under these conditions, the water molecules
47 increase their mobility. Nevertheless, an increase in temperature may affect compounds
48 which are of interest.

49 The air drying velocity is another important process condition. It influences the
50 external resistance to heat and mass transfer. Thus, if the external resistance is negligible,
51 an increase in the air drying velocity will not influence the drying kinetics. For that

52 reason, from an energy point of view, it is important to establish a threshold value for air
53 velocity when a particular product is dehydrated^[6, 7].

54 Another process condition which may be taken into account is the relative humidity
55 of the drying air. This parameter, together with temperature, determines the drying
56 potential of the air. When the air has a high temperature and a low relative humidity, its
57 drying potential will be high. In convective drying, room air is heated until it reaches the
58 temperature sought for the drying process. In this operation, the drying potential of air is
59 increased because its temperature rises and its level of relative humidity falls.

60 Recently, the application of high intensity ultrasound during air drying has been
61 considered as an intensification technology. Ultrasound is mainly applied in food
62 processes due to the effects it exerts on heat or mass transfer operations^[8]. Ultrasonic
63 waves are transmitted in a relatively easy way in liquids. For that reason, most of the
64 ultrasonic applications reported in literature are found in liquid–liquid and liquid–solid
65 systems^[9]. Some examples of these applications can be found in the literature^[8]. In gas-
66 solid systems, like convective air drying, the high impedance mismatch and the high
67 ultrasonic energy attenuation in air make the transmission of ultrasound from the
68 transducer to the air and from the air to the solid difficult^[10]. Nevertheless, some studies
69 can be found in literature into high intensity ultrasound-assisted convective air drying^{[10,}
70 ^{11, 12, 13, 14]}. In some cases, it has been found that over 70% less time is needed for the
71 drying process^[14]. Nevertheless, as the effects of ultrasounds are product and drying
72 variables (temperature, air velocity) dependent, the drying gains should be considered
73 case by case. So far, all this research is laboratory scale and no ultrasound application has
74 been found in the industrial drying of food products.

75 Power ultrasound assisted convective drying (acoustic drying) may constitute a
76 means of improving the dehydration rate without significantly heating the material^[8].

77 The application of ultrasound during convective air drying increases the kinetics of
78 dehydration, affecting both the internal and the external resistance^[8]. As regards internal
79 resistance, when ultrasound travels across a medium, it produces alternating cycles of
80 expansions and contractions (sponge effect), helping the water to leave easily and
81 diminishing the internal resistance to mass transfer^[15]. These effects are product
82 dependent, and the texture of the material is a key parameter. Ozuna et al.^[16] found that
83 the sponge effect was more intense in soft products. It seems that the expansions and
84 contractions would be diminished in hard products due to the fact that their solid matrix
85 moves with difficulty.

86 The influence of ultrasound on the external resistance to mass transfer could be
87 linked to the generation of differential pressures and the microstirring at the interfaces^[8].
88 Although these effects are not observed, if internal resistance prevails, as consequence no
89 change will be found in the mass transfer coefficient identified.

90 It is essential to model the drying kinetics in order to carry out the engineering design
91 of the drying processes and evaluate the effects of the variables considered.

92 In the literature, research can be found into modeling the drying of different grain
93 food, for example, soya bean^[17], rice^[18] or corn^[19]. These models can be theoretical or
94 empirical. The theoretical models, for example the diffusion model, are based on the
95 understanding of the phenomenon under study, whereas the empirical models, for
96 example Peleg's model, are based on empirical approximations which are product of the
97 observation or experimentation. For that reason, theoretical models are more complicated
98 from a mathematical point of view. Empirical models are easy to solve and sometimes the
99 results provided are good enough for the purpose sought. In general, the simplest model is
100 always recommended in order to facilitate its solution and use in real time for control or
101 optimal operation management^[7].

102 In the literature, there is scarce research into the influence of the drying conditions on
103 the dehydration process of grape seeds and the effect of ultrasounds on a hard product is
104 lacking. Thus, the aim of this work was to determine the influence of temperature, air
105 velocity and the application of ultrasounds on the drying kinetics of grape seeds, in order
106 to provide a sound basis for industrial process management.

107

108 **MATERIALS AND METHODS**

109 **Raw material**

110 Grape pomace from the wine processing of red grapes (*Vitis vinifera* var Bobal) was
111 collected from a winery located in Requena (Valencia, Spain). It was packed in a plastic
112 film, in order to avoid moisture loss until its constituents were separated, and refrigerated
113 at 2 ± 0.2 °C. The separation of skins, seeds and stems was performed manually. After
114 separation, grape seeds were again plastic wrapped and refrigerated at 2 ± 0.2 °C until the
115 determination of the drying kinetics.

116 The initial moisture content of grape seeds was determined by drying them at 70 °C
117 under vacuum conditions until constant weight was reached^[20].

118

119 **Determination of experimental drying kinetics**

120 The experimental drying kinetics were obtained in triplicate at 1.0, 1.5, 2.0 and 3.0
121 m/s air velocity and 40, 50, 60 and 70°C air temperature. All these temperatures were
122 under the smoke point of the grape seed oil^[4]. The relative humidity at the air inlet for all
123 the drying kinetics was 71.4 ± 6.0 %. When the air was heated, this value falls to $15.0 \pm$
124 8.9 %. In order to determine the influence of ultrasound application, the drying kinetics
125 were obtained in triplicate at 1.0 and 1.5 m/s with and without ultrasound application
126 (30.8 kW/m^3). When ultrasounds were applied, their intensity measured as Sound
127 Pressure Level was 154.1 dB and the electroacoustic efficiency was around 60-70 %. For

128 experimental drying kinetics determination, the convective dryer described previously by
 129 Cárcel at al.^[11] was used (Figure 1). This equipment is a pilot scale convective dryer
 130 modified to apply power ultrasound. The dryer has remote control temperature and air
 131 velocity and the weight of the sample was monitored periodically during the drying
 132 period. Grape seeds were placed into the drying chamber on a perforated parallel plate
 133 support, in order to guarantee that the hot air affects the entire surface of the seeds. In each
 134 experiment, 22.3 ± 2.3 g of grape seeds were used. The drying was carried out in
 135 stationary bed.

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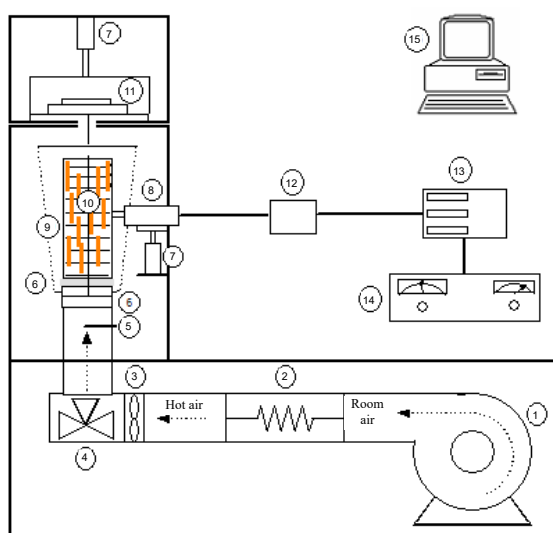
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146 FIG. 1. Convective dryer. (1) Fan; (2) Heating; (3) Anemometer; (4) Pneumatic three-way
 147 valve; (5) Temperature probe Pt-100; (6) Coupling material; (7) Elevator; (8) Ultrasonic
 148 transducer; (9) Braces; (10) Parallel plate support; (11) Scale; (12) Impedance matching
 149 unit; (13) Digital wattimeter; (14) Generator of power ultrasound; (15) Computer-
 150 controller.

151

152

153

154 **Modelling of experimental drying kinetics**

155 Two models were used to model the experimental drying kinetics: Peleg's model and
156 a simplified model based on Fick's second law.

157

158 *Peleg's model*

159 Peleg's model^[21] has been used satisfactorily to model the dehydration of grain food
160 products^[22]. It is shown in equation 1.

$$161 \quad X = X_0 - \frac{t}{k_1 + k_2 t} \quad (1)$$

162 The constant, k_1 , is a kinetic parameter. Sopade et al.^[23] proposed describing the
163 effect that temperature has on k_1 by means of the Arrhenius equation and the activation
164 energy can be assessed as follows (equation 2).

$$165 \quad \frac{1}{k_1} = \frac{1}{k_p} \exp\left(\frac{-E_a}{\mathfrak{R}T}\right) \quad (2)$$

166 From equation 2, the activation energy can be identified from the slope of the fitted
167 straight line by plotting $1/k_1$ versus $\ln(1/T)$.

168

169

170 *Difusion model*

171 As no constant drying rate period was observed, the initial moisture content and
172 critical moisture content were considered equal; thus, only the falling drying rate was
173 considered in the model^[24]. Shrinkage and external resistance were not considered in this
174 model. Seeds were considered to be homogeneous, isotropic and spherically shaped. The
175 governing equation (equation 3), the initial equation (equation 4) and the boundary
176 conditions (equations 5 and 6) are shown:

$$177 \quad \frac{\partial X_l}{\partial t} = D_e \left(\frac{\partial^2 X_l}{\partial r^2} + \frac{2}{r} \frac{\partial X_l}{\partial r} \right) \quad (3)$$

178
$$X_l(r,0) = X_0 \quad (4)$$

179
$$X_l(R,t) = X_e \quad (5)$$

180
$$\frac{\partial X_l}{\partial r}(0,t) = 0 \quad (6)$$

181 The equilibrium moisture content was calculated by means of Peleg's model
182 (equation 7).

183
$$X_e = X_0 - \left(\frac{1}{k_2} \right) \quad (7)$$

184 The diffusion model was solved by using the method of the Separation of Variables
185 and the effective diffusivity was identified by means of an optimization method^[25,24]. The
186 objective function was the squared differences between the experimental and calculated
187 values of the average moisture content. The minimization of the objective function was
188 performed using the tool Solver from Microsoft Excel.

189 The influence of temperature on the effective diffusivity was assessed by means of
190 the Arrhenius equation (equation 8).

191
$$D_e = D_0 \exp\left(\frac{-E_a}{\mathfrak{R}T}\right) \quad (8)$$

192

193 *Evaluation of the quality of fit*

194 The goodness of fit for the two models was assessed by means of the explained
195 variance^[26].

196 Additionally, both a t-test and a Lilliefors test were performed at 1 the 5%
197 significance level for both models and also for the Arrhenius equation. The t-test served to
198 evaluate whether the data in the residual vector are random and have a normal distribution
199 with mean 0 and unknown variance, against the alternative that the mean is not 0. The
200 Lilliefors test was used to test the assumption that the residual vector comes from normal

201 distributions. The “ttest” function and “lillietest” function of the software Matlab®
202 R2011^[27] were used to perform the t-test and the Lilliefors test, respectively.

203 The result of the t-test was a confidence interval (C_i). There was a 95% probability of
204 the residual vector mean being in the confidence interval.

205 In the Lilliefors test, the statistical value (kstat) and the critical value (critval) were
206 the results. If kstat was lower than critval, there was a 95% probability of the normality of
207 the residuals being established.

208

209 RESULTS AND DISCUSSION

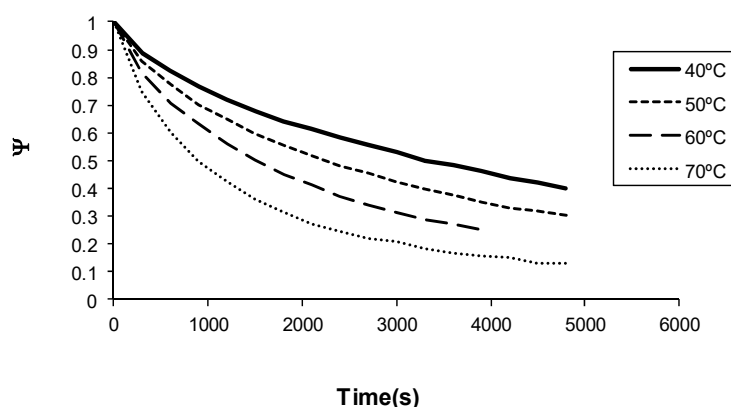
210 Experimental drying kinetics

211 In Figures 2, 3 and 4 some experimental drying kinetics are plotted in order to
212 illustrate the effect of temperature, air velocity and ultrasound application. Each of the
213 represented experimental drying kinetics is the average of the three replications for each
214 of the experimental conditions. The time interval measurements were the same for all the
215 experiments, which allowed the data obtained on the three replicates to be averaged.

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225 FIG. 2. Experimental drying kinetics at different temperatures and an air velocity of 1 m/s
226 (without ultrasound application)

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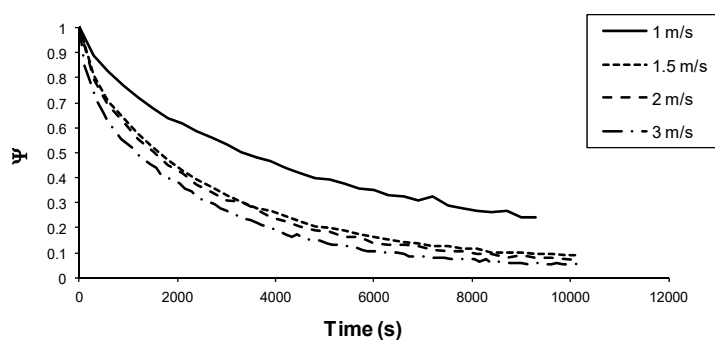
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235 FIG. 3. Experimental drying kinetics at different air velocities and a temperature of 40°C
 236 (without ultrasound application)

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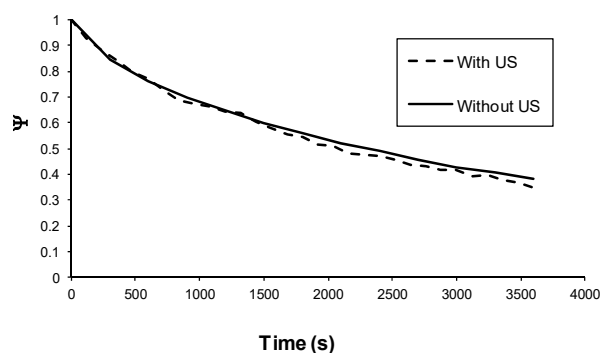
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244 FIG. 4. Experimental drying kinetics at 40 °C and 1m/s with and without ultrasound
 245 application

246

247 As can be observed in Figure 2, in the range considered, when the temperature rises
 248 the drying kinetics also increases. This result coincides with what has been found in other
 249 studies^[28,29].

250 The influence of drying air velocity is assessed in Figure 3. When the air velocity
 251 increases from 1 m/s to 1.5 m/s, less time is needed to attain a given moisture content.
 252 Nevertheless, for an air velocity of between 1.5 and 3 m/s, this parameter is only observed
 253 to have a slight influence on drying kinetics. It seems that the external resistance to mass
 254 transfer is negligible for an air velocity of over 1.5 m/s. Similar results have been found

255 by other authors when studying different agro-food products: broccoli^[30], turmeric^[6] or
256 meat^[7], where different velocity thresholds were observed depending on the product.

257 Figure 4 shows that, under the experimental conditions 1 in this study, there was no
258 influence of ultrasound application on the dehydration kinetics.

259

260 **Modelling**

261 Modelling was carried out for all the drying kinetics separately and, after that, the
262 average and standard deviations were calculated for the parameters obtained from each
263 model.

264 Table 1 shows the results for the parameters of Peleg's model. The percentage of
265 explained variance for all the drying kinetics was over 98%, thus the agreement between
266 the experimental and calculated values can be considered a good one. As regards the t-
267 test, the 0 is contained in every confidence interval for all the drying kinetics. As to the
268 Lilliefors test, the statistical value is lower than the critical value for all the experimental
269 conditions. Thus, the residuals followed a normal distribution and their mean was 0, with
270 a significance level of 5%. As an example, in Figure 5 a comparison between two
271 experimental drying kinetics and Peleg's model results are shown. The same behavior was
272 observed for all the other drying conditions.

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280 TABLE 1: Results for the parameters of Peleg's model, sd (standard deviation), var
 281 (explained variance)

Sample	$k_1 \pm \text{sd}$	$k_2 \pm \text{sd}$	var (%)
1 m/s without US			
40 °C	4470 ± 61	2.09 ± 0.21	99.6
50 °C	3006 ± 85	1.90 ± 0.52	90.7
60 °C	2100 ± 49	1.77 ± 0.01	99.5
70 °C	1818 ± 68	1.55 ± 0.25	97.7
1.5 m/s without US			
40 °C	4049 ± 72	1.79 ± 0.32	99.5
50 °C	2450 ± 172	1.70 ± 0.02	95.0
60 °C	1652 ± 37	1.51 ± 0.16	99.8
70 °C	1328 ± 136	1.46 ± 0.02	95.8
2 m/s without US			
40 °C	3424 ± 149	1.81 ± 0.11	99.7
50 °C	2317 ± 85	1.72 ± 0.03	99.8
60 °C	1427 ± 115	1.58 ± 0.02	99.7
70 °C	1162 ± 9	1.42 ± 0.03	90.1
3 m/s without US			
40 °C	2646 ± 48	1.96 ± 0.12	99.8
50 °C	1496 ± 45	1.80 ± 0.01	99.9
60 °C	1137 ± 78	1.61 ± 0.02	91.4
70 °C	843 ± 122	1.50 ± 0.11	99.9
1m/s with US			
40 °C	4400 ± 40	1.70 ± 0.19	95.5
50 °C	3018 ± 28	1.66 ± 0.15	99.4
60 °C	2076 ± 20	1.61 ± 0.14	99.2
70 °C	1789 ± 24	1.46 ± 0.10	98.1
1.5 m/s with US			
40 °C	3792 ± 195	1.60 ± 0.14	98.6
50 °C	2152 ± 249	1.56 ± 0.01	98.8
60 °C	1707 ± 31	1.51 ± 0.14	98.7
70 °C	1117 ± 102	1.45 ± 0.09	99.6

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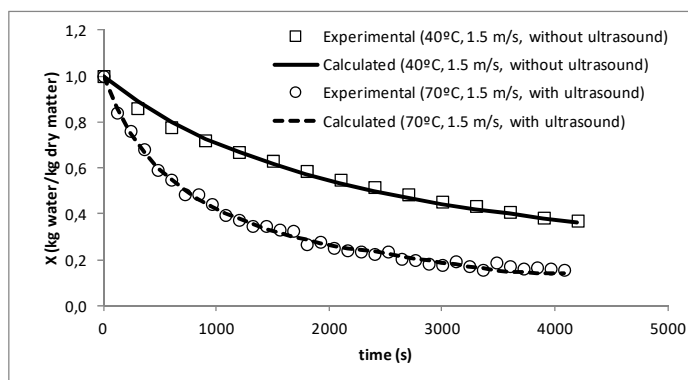
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293 FIG. 5. Comparison between modeled by Peleg's model and experimental drying curves
 294 for two drying conditions (40°C, 1.5 m/s, without ultrasound and 70°C, 1.5 m/s, with
 295 ultrasound)

296

297 As expected, the kinetic parameter k_1 decreased when the temperature rose. It can
 298 also be observed that the values for k_1 were similar regardless of whether ultrasounds
 299 were applied or not. Thus, it seems that the application of ultrasounds has no influence on
 300 the drying kinetics under the drying conditions considered in this study.

301 For all the drying kinetics, the equilibrium moisture content calculated by means of
 302 Peleg's model was 0.05 ± 0.02 kg water/kg dry matter. This low equilibrium moisture
 303 content value indicates that the relative humidity of the drying air did not influence the
 304 drying kinetics.

305 After modeling the experimental results by means of the diffusion model, the results
 306 shown in Table 2 were obtained. There is a good agreement between the experimental and
 307 calculated values. The explained variance was higher than 90.5% for all the drying
 308 kinetics and the residuals followed a normal distribution, with a mean of 0 and a
 309 significance level of 5%.

310

311

312 TABLE 2: Effective diffusivity (D_e), standard deviation (sd) and explained variance (%)
 313 var) for the different drying conditions

314

1 m/s				
With US			Without US	
Temperature	$(D_e \pm sd) \cdot 10^{10}$ m ² /s	% var	$(D_e \pm sd) \cdot 10^{10}$ m ² /s	% var
40 °C	0.55±0.02	95.5	0.51±0.04	96.0
50 °C	0.78±0.03	93.2	0.75±0.08	90.7
60 °C	1.18±0.12	95.8	1.19±0.04	91.0
70 °C	1.56±0.14	92.9	1.48±0.23	90.8
1.5 m/s				
With US			Without US	
Temperature	$(D_e \pm sd) \cdot 10^{10}$ m ² /s	% var	$(D_e \pm sd) \cdot 10^{10}$ m ² /s	% var
40 °C	0.68±0.04	96.6	0.65±0.08	95.3
50 °C	1.08±0.04	94.2	1.01±0.05	93.7
60 °C	1.55±0.04	96.3	1.43±0.12	95.5
70 °C	1.87±0.05	95.2	1.85±0.13	92.3
2 m/s			3 m/s	
Without US			Without US	
Temperature	$(D_e \pm sd) \cdot 10^{10}$ m ² /s	% var	$(D_e \pm sd) \cdot 10^{10}$ m ² /s	% var
40 °C	0.71±0.05	93.8	0.78±0.06	91.6
50 °C	1.17±0.13	94.1	1.18±0.13	91.7
60 °C	1.52±0.06	94.7	1.57±0.10	96.9
70 °C	1.89±0.03	93.0	1.87±0.03	93.0

315

316 Figure 6 represents an example of the comparison between calculated by means of
 317 diffusion model and experimental drying curves for two experimental drying conditions.

318 The agreement between experimental and calculated values was good for all the drying
 319 conditions considered.

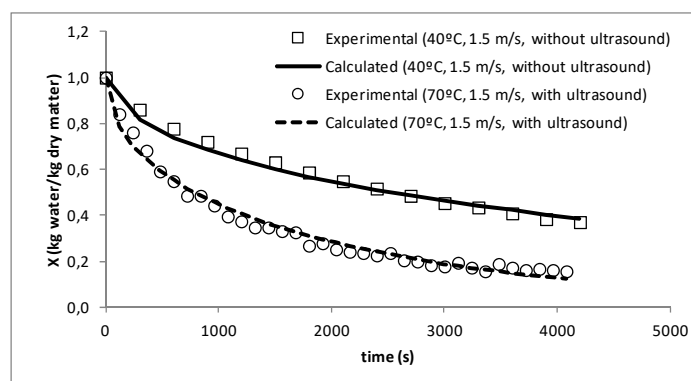
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330 FIG. 6. Comparison between modeled by diffusion model and experimental drying
331 curves for two drying conditions (40°C, 1.5 m/s, without ultrasound and 70°C, 1.5 m/s,
332 with ultrasound)

333

334 The effective diffusivity values obtained are in the range reported by Saravacos and
335 Maroulis^[31] for agro-food products, which is between $1 \cdot 10^{-11}$ and $1 \cdot 10^{-8}$. The values also
336 coincide with others found in literature for grape seeds^[29].

337

338 **Influence of temperature**

339 As expected, there was an influence of the temperature on k_1 and D_e as can be seen
340 in Tables 1 and 2, respectively. This influence was assessed in terms of the Arrhenius
341 equation (equation 2 for Peleg's model and equation 8 for the diffusion model). Table 3
342 shows the activation energy and the pre- exponential values obtained. Regarding to t-test
343 and Lilliefors test, the residuals followed a normal distribution, with a mean of 0 and a
344 significance level of 5% when the Arrhenius equation was used for the calculation of E_a
345 from both models (the diffusion and Peleg's). Figure 7 shows the values of k_1 calculated
346 by the Arrhenius equation versus k_1 from Peleg's model. The fit between the D_e from the
347 diffusion model and from the Arrhenius equation is represented in Figure 8. The
348 correlation coefficient was 0.99 and 0.97 respectively; thus, the Arrhenius equation
349 satisfactorily described the influence of temperature on the drying kinetics.

350 TABLE 3: Activation energy (E_a , kJ/mol) obtained by means of both models considered.
 351 CI (Confidence Interval, 95%). Pre-exponential factors, D_0 (diffusion model) and k_p
 352 (Peleg's model)

		Peleg's model			Diffusion model		
		$E_a \pm CI$ (kJ/mol)	k_p	var (%)	$E_a \pm CI$ (kJ/mol)	D_0 (m^2/s)* 10^5	var (%)
1 m/s	with US	27.8 ± 4.3	0.094	97.1	31.6 ± 4.9	1.02	95.9
	without US	27.4 ± 5.1	0.113	96.7	32.2 ± 8.6	1.25	91.8
1.5 m/s	with US	34.8 ± 8.2	0.005	94.8	30.3 ± 4.1	0.83	96.8
	without US	33.0 ± 5.6	0.012	95.1	31.8 ± 6.0	1.36	94.1
2 m/s	without US	34.0 ± 4.7	0.007	96.7	28.8 ± 5.4	0.49	93.3
3m/s	without US	33.5 ± 5.8	0.006	94.3	24.4 ± 5.2	0.10	91.5

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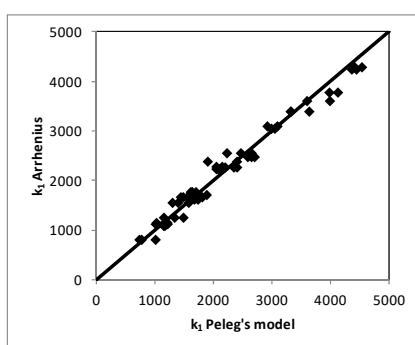
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359 FIG. 7. Values of k_1 calculated by Arrhenius equation versus k_1 from Peleg's model for
 360 all the temperatures under study

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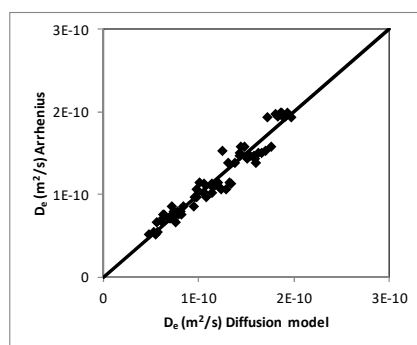
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367 FIG. 8. Values of D_e calculated by Arrhenius equation versus D_e from diffusion model
 368 for all the temperatures under study

369 As can be observed in table 3, the E_a values were not significantly influenced by air
 370 velocity. According to Ramallo et al.^[32], the activation energy values for agro-food
 371 products range from 15 to 95 kJ/mol. Thus, the values in table 3 are in the range reported
 372 by these authors. They are also similar to the values found in literature for some products,
 373 for example 30.45 kJ/mol for grape seeds^[29] or 30.37 kJ/mol for aloe vera^[33].

374 No significant differences can be observed between the values of E_a given by Peleg's
 375 model or those provided by the diffusion model. Using Peleg's model to calculate the
 376 activation energy offers the advantage that it is easier from a mathematical point of view
 377 and, as can be seen in Table 3, the results are comparable with those obtained using the
 378 diffusion model.

379

380 **Influence of air velocity**

381 Figure 9 shows the influence that air velocity has on effective diffusion coefficient at 70°C
 382 (no ultrasound application). Similar results were obtained for the other temperatures under
 383 study.

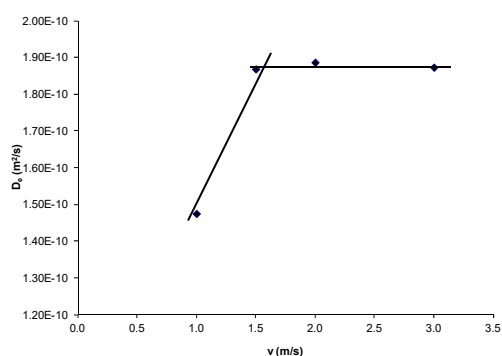
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389 FIG. 9. Influence of air velocity on drying kinetics at 70°C (no ultrasound application)

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391 For an air velocity between 1.0 and 1.5 m/s, an increase in this parameter leads to a
 392 significant increase in effective diffusivity. For an air velocity of over 1.5 m/s, the
 393 effective diffusivity is not affected by air velocity. The diffusion model considered in this
 394 research work did not take into account the external resistance to mass transfer; thus,

395 calculated D_e included both resistances to mass transfer (internal and external). The effect
396 of an air velocity of between 1.0 and 1.5 m/s on D_e can be attributed to the influence of
397 external resistance to mass transfer for these air velocities. If external resistance is not
398 borne in mind when it is important, the values of D_e can be underestimated^[7]. Thus,
399 Figures 3 and 9 seem to indicate that for an air velocity of over 1.5 m/s, the external
400 resistance to mass transfer is negligible when grape seeds are dehydrated. This threshold
401 matches others found in literature^[34,5,7].

402 Because the models considered described the drying kinetics reasonably well, they
403 can be used to analyze the effects of ultrasounds.

404

405 **Influence of ultrasound application**

406 Tables 1 and 3 show that, at a particular temperature and for a specific air velocity,
407 there is no significant difference between parameters k_1 (Table 1) and D_e (Table 3)
408 obtained for drying kinetics with and without ultrasound application. The activation
409 energy (E_a) was not influenced by ultrasound application either.

410 Ozuna et al.^[16] have identified a relationship between the textural properties of
411 vegetables and how the drying process is affected by the application of ultrasound at 40°C
412 and 1 m/s. This effect was dependent on the ultrasonic power applied: the higher the
413 power, the larger the identified effective diffusivity. Nevertheless, the improvement
414 brought about by the effect of ultrasound on the effective diffusivity was closely
415 correlated with the hardness of the product. These authors found that, when ultrasounds
416 were applied over a wide power range, they only exert a slight influence on the drying
417 kinetics in vegetable products with high levels of hardness. Working on lemon peel and
418 carrot, García-Pérez et al.^[10] [10] dehydrated both products at 40 °C and 1 m/s and found
419 that for the former any power of ultrasound influenced the drying kinetics, whereas for
420 the latter, this influence was detected only from a threshold power value. It seems that

421 ultrasound is less effective on the internal resistance of hard products. Thus, the
422 mechanical compressions and expansions (“sponge effect”) produced by ultrasound
423 application, which enhanced the water removal, were more intense in soft products.

424 According to Milani et al.^[35], the hardness of the grape seeds is 45.83 N, a higher
425 value than for carrot^[16]. For vegetable products with this level of hardness, ultrasound
426 application should show no influence on drying kinetics when applied at 30.8 kW/m³^[16],
427 which is the maximum power of the equipment used in this research. Thus, the high level
428 of hardness of the grape seeds would explain the fact that ultrasound has no influence on
429 the drying kinetics under the experimental conditions considered.

430 Figure 9 shows that, at an air velocity of under 1.5 m/s , external resistance affects
431 drying kinetics. As a consequence, ultrasounds should enhance drying kinetics by
432 affecting external resistance. Nevertheless, this was not the case; this could be linked to
433 the interaction of acoustic energy with the product. Apparently, there is a large reflection
434 of the acoustic energy impinging on the product, which could be due to the great
435 mismatch of acoustic impedance (air – seed) and to the smooth external layer of the seeds.
436 This effect merits further investigation. Apparently, there is not only a phenomena linked
437 to applied power, but also to product characteristics.

438

439 CONCLUSION

440 The experimental drying kinetics of grape seeds were modelled using both Peleg’s
441 model and the diffusion model. The results obtained are similar to others in literature.
442 There was an observed increase in the drying kinetics when the temperature rose. The
443 activation energy was calculated by means of the two models considered. The value
444 ranged between 27.4 and 34.8 kJ/mol. No significant differences in the values of E_a were
445 found for any of the drying conditions considered. Peleg’s model had the advantage of
446 being mathematically simple and, consequently, useful for real-time applications.

447 For an air velocity of 1.5 m/s or higher, the external resistance 1 to mass transfer does
 448 not influence the dehydration process under the experimental conditions used in this
 449 research. As a consequence, in order to save energy, this threshold should not be
 450 exceeded.

451 Under the experimental conditions considered, ultrasound application had no
 452 influence on the dehydration kinetics of grape seeds. This may show that the physical
 453 characteristics, hardness and low porosity, of the grape seeds may influence the reflection
 454 of the acoustic waves reaching the products. This should be investigated further.

455

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459

460 **NOMENCLATURE**

461	D_e	effective diffusivity, m^2s^{-1}
462	D_0	pre-exponential factor, m^2s^{-1}
463	E_a	activation energy, $kJmol^{-1}$
464	k_1	Peleg’s model parameter, $s(kg\ water/kg\ dry\ matter)^{-1}$
465	k_2	Peleg’s model parameter, $(kg\ water/kg\ dry\ matter)^{-1}$
466	k_p	pre-exponential factor
467	t	time, s
468	T	temperature, K
469	X	mean moisture content, db
470	X_{cal}	calculated mean moisture content, db
471	X_e	equilibrium moisture content, db
472	X_{exp}	experimental mean moisture content, db
473	X_l	local moisture content, db
474	X_0	initial moisture content, db
475	r	length co-ordinate, m
476	R	radius of the seeds, m
477	\mathfrak{R}	constant of perfect gases (8.31), $JK^{-1}mol^{-1}$
478	Ψ	dimensionless moisture content, $\Psi = (X - X_e)/(X_0 - X_e)$
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