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Additional Information

INTENSIFICATION OF LOW TEMPERATURE DRYING BY USING ULTRASOUND

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Abstract

Low temperature air drying involves temperatures below or close to the freezing point and aims to reduce the water content or to remove organic solvents keeping quality attributes, thus it has a great potential in food, chemical, pharmaceutical and cosmetic industries. Depending on the temperature and solvent involved, the removal is by evaporation or sublimation, but in all cases, the drying process is slow due to the low temperature used. An efficient ultrasound application at atmospheric pressure and moderate temperatures could accelerate the drying process. Thus, the main aim of this work was to test the feasibility of power ultrasound to intensify low temperature drying processes.

Drying kinetics of carrot, eggplant and apple cubes (side 10 mm) were carried out at atmospheric pressure, 2 m/s, -14 ºC and relative humidity 7% with (acoustic power 19.5 kW/m^3) and without ultrasound application. At the same experimental conditions, kinetics of ethanol removal from a solid matrix were also performed. Diffusion models were used to describe drying curves and identify kinetic parameters in order to evaluate and quantify the process intensification attained by ultrasound application.

The effect of ultrasound application was similar for all products tested, being drying time shortened between 65-70 %. In the case of ethanol removal, the time reduction by ultrasound application was even higher achieving 120 %. Both, effective moisture diffusivity and mass transfer coefficient increased from 96 to 170 % and from 407 to 428 % when ultrasound was applied, respectively. Therefore, ultrasound application should be considered a potential and effective technology to intensify low temperature drying processes, being capable to make more affordable and less energy and time-consuming these processes for all kind of industries.

Keywords: ultrasonic, atmospheric freeze drying, diffusion, mass transfer, solvent removal.

1. Introduction

The removal of solvents at low temperature is considered a common stage in food, chemical, pharmaceutical and cosmetic industries (Claussen et al., 2007; Kudra and Mujumdar, 2009; Reyes et al., 2010). Processing involves temperatures below or close to the freezing point and aims to reduce the water content or to remove organic solvents keeping quality attributes. Thus, vacuum freeze drying of pharmaceuticals (Andrieu and Vessot, 2011), atmospheric freeze drying of fish in northern Europe or drying process of dry-cured meat products (Gou et al., 2002) are some instances of low temperature drying processes. Depending on the temperature and solvent involved, the removal is by evaporation or sublimation, but in all cases, involves long drying times. As an illustration, the drying time in the processing of dry-cured ham ranges from 4 month to over 2 years (Gou et al., 2002). Therefore, there exists a great opportunity for process intensification, which is a growing trend in process engineering to achieve more sustainable and affordable technologies.

Process intensification aims at the improvement of traditional technologies and the development of new technologies to reach higher yield, notable reduction in equipment costs, lower energy use and increase product quality and processing safety (Benali and Kudra, 2010). Traditionally, drying at low temperature has been intensified by working at vacuum conditions, which is referred to the well known conventional freeze drying, which provides high quality products and where water removal occurs mainly by sublimation and product keeps frozen. Freeze drying has high fix and operational costs (Claussen et al., 2007b) due to high energy consumption and demanding requirements of vacuum equipments and requires batch production, which makes a costly and exclusive process. Process intensification can be also addressed by coupling new technologies to atmospheric or convective freeze drying, which mainly consists of flowing air at low temperature (below freezing point) to the product being dried providing dry products with similar quality than conventional freeze dried ones (Reyes et al., 2010). In such a way, the product also keeps frozen during drying and water removal occurs mainly by sublimation. The introduction of new technologies with high heating effect, such as microwave, radio-frequency and infrared radiation, should be avoided if possible due to the risk of overheating and product thawing with the subsequent reduction of the dry product quality (Riera et al., 2011). Therefore, to avoid this risk without a costly strict control of the process, non thermal strategies are mostly required for this process intensification, like the application of power ultrasound.

The use of power ultrasound has been recently explored for conventional hot air drying of different vegetables and fruits (Gallego-Juarez et al., 1999; Mulet et al., 2003; Garcia-Perez et al., 2011b and Riera et al., 2009). More efforts have been addressed to design and develop efficient ultrasonic devices (Gallego-Juarez et al., 2007) due to the complex application of power ultrasound in gas media. From the previous studies, it was concluded that an efficient power ultrasound application produce mechanical effects both on the gas-solid interfaces and in the material being dried. Thus, ultrasound may intensify water removal without introducing a high amount of thermal energy during drying (Riera et al., 2011). Therefore, the use of power ultrasound either to dry heat sensitive materials or to be applied in low-temperature drying processes has great potential that needs to be investigated (Garcia-Perez, 2007). Due to the ultrasound application being dependent on process and product variables, such as temperature, air velocity, acoustic power applied and product porosity, it results necessary to evaluate its feasibility when a new use is addressed.

Due to the lack of evidence on the efficient use of ultrasound on drying at low temperature, it is interesting to assess the feasibility of an efficient power ultrasound application to intensify this process. The drying process of vegetables products of different porosity at temperatures below freezing point will be addressed as well as the removal of other solvents from solid matrixes. Modeling was aimed at quantifying the influence of power ultrasound on mass transfer processes, although more insight will be needed in the future by considering more accurate mechanistic models.

2. Materials and methods

2.1. Ultrasonically assisted air drier

Drying experiments were carried out in a convective drier with air-recirculation, temperature and air velocity control and an ultrasonically activated drying chamber, a schematic layout of this system is shown in Fig. 1.

Air flow is driven by a medium pressure fan (COT-100, Soler & Palau, Spain) and measured by a van anemometer (1468, Wilh. Lambrecht GmbH, Germany). The air velocity (from 0.1 to 20 m/s) was controlled by a PID algorithm by using a digital inverter (MX2, Omron, Japan) that acts over the fan speed. In order to reach low temperatures in the air flow, a copper tube heat exchanger (area 13 m^2, fin space 9 mm, Frimetal, Spain) using a glycol/water solution (45 %, v/v) was installed in the air duct. A chiller (MTA, Italy) placed out of the drier provided the glycol/water solution at -22 ºC and 150 L/min. The air temperature and relative humidity was measured in three points of the air duct (KDK, Galltec+mela, Germany): drying chamber inlet and heat exchanger inlet and outlet. Air temperature (from 60 to -10 ºC) was also controlled by a PID algorithm acting over the electrical heating elements (maximum power 2500 W, 230 V). In the temperature control system, a Pt-100 probe was used due to its shorter response time than combined air and relative humidity sensors. A compact Field Point (cFP-2220, National Instruments, USA) with RTD, analog and digital input and output modules was used to supervise and create the control loops of the air velocity and temperature.

In order to control the relative humidity, air flow was forced to go through 3 trays of silica gel. Each day, one tray was substituted to be re-generated (150 ºC).

A high-power ultrasonic application system already described in previous works was assembled in the new convective drier to be used as drying chamber (Garcia-Perez et al., 2006). It mainly consists of a cylindrical radiator driven by a power ultrasonic transducer (frequency 21.9 kHz, impedance 369 Ω, power capacity 90 W). Ultrasonic signal is generated and fitted to minimize the phase between electric voltage and intensity by a resonance dynamic controller (PUSONICS, Spain), while the power capacity is kept by acting over the power amplifier (PUSONICS, Spain). Finally, an impedance matching unit (impedance from 50 Ω to 500 Ω and inductance from 5 and 9 mH, PUSONICS) is used to electrically optimize the ultrasonic application. The ultrasonic system provides an average sound pressure level in the drying chamber of 155 dB. The resonance dynamic controller was connected to a PC by the RS-232 interface to adequately monitor the main electric properties applied during the ultrasonic application (Power, Intensity, Voltage, Phase, Frequency and Impedance).

Air flow goes through the cylindrical radiator where samples are placed. In order to determine the drying kinetics, samples are weighted at preset times by using an industrial weighing module (6000±0.01 g, VM6002-W22, Mettler-Toledo, USA) connected to the compact Field Point by the RS-422 interface. A weighing sequence was programmed in the compact Field Point to make an accurate measurement: the fan was stopped and the electric slide table actuator (LEC 6, SMC, Japan) moved the samples outside the cylindrical radiator to take the weight. Thereby, the noise introduced by vibration of the cylindrical radiator in the weighing unit was avoided.

An application was developed using LabVIEW2009™ programming code (National Instruments, USA) to make an overall control and monitoring of the ultrasonically intensified drying process, integrating air flow, sample and ultrasonic parameters information. The application can indistinctly run in either the PC or compact Field Point.

2.2. Drying experiments

Vegetal material with different internal structure was used in the drying experiments. Thus, low (carrot, var. Nantesa), medium (apple, var. Granny Smith) and high (eggplant, var. Black Enorma) porosity products (Boukouvales et al., 2006) were purchased in local markets. For all the products, cubic samples (side 10 mm) were obtained from the flesh using a commercial slicing system (CL50E, Robot Coupe, France). Samples were wrapped using plastic and frozen by placing in a freezing room at -20 ºC until processing. In all cases, storage time was shorter than 72 hours. Initial moisture content was measured according to AOAC method nº 934.06 (AOAC, 1996).

Air drying (AIR) experiments were carried out at -14±1 ºC and 2±0.1 m/s and an average relative humidity of 7±3 %. Ultrasonically assisted air drying experiments (AIR+US) were carried out in the same experimental conditions applying an acoustic power of density 19.5 kW/m^3, which is defined as the electric power applied to the ultrasonic transducer (45 W) divided by the volume of the drying chamber (cylindrical radiator, 2.3 L). Initial mass load densities ranging between 6 (for eggplant) and 10.5 kg/m^3 (for carrot) were used. Samples were weighed at preset times ranging between 1 and 5 hours. Sample weight losses of 90 (eggplant), 85 (carrot) and 83 % (apple) were set to finalize the drying experiments, which were replicated, at least, three times for all conditions.

In order to test the ability of ultrasound to remove other solvent different to water from solid matrix, additional drying experiments were carried out. AIR dried apple samples were impregnated with ethanol (96 % v/v) at vacuum conditions (absolute pressure 0.3 atm) for 60 min. Afterwards, ethanol removal experiments were carried out without (AIR) and with (AIR+US) ultrasound application at the same experimental conditions than air drying ones, -14±1 ºC, 1±0.1 m/s and 19.5 kW/m^3. Ethanol removal kinetics were determined by weighting samples at preset times. Experiments were replicated, at least, three times.

2.3. Modeling

The aim of this study being the feasibility of the use of ultrasound, thus, modeling was considered to quantify its effects by using a simplified model where water loss during drying was described by considering the diffusion theory. Eq. 1 shows the mass transfer governing equation obtained by considering a uniform temperature, constant effective diffusivity and negligible shrinkage.

$$\frac{\partial W_p(x,y,z,t)}{\partial t} = D_e\left(\frac{\partial^2 W_p(x,y,z,t)}{\partial x^2} + \frac{\partial^2 W_p(x,y,z,t)}{\partial y^2} + \frac{\partial^2 W_p(x,y,z,t)}{\partial z^2}\right) \quad \text{(Eq. 1)}$$

where W_p is the averag water content (kg water/kg dry matter, dm), D_e is the average effective diffusivity (m^2/s), t time (s) and x, y, z represent characteristic coordinates of cubic geometry.

In order to solve Eq. 1., uniform initial water content was assumed as the initial condition. As boundary conditions, solid symmetry was considered in x, y and z mass transfer directions. Two different approaches related to convection were taken into consideration. As first approach, a negligible boundary layer thickness was assumed, thus, surface water content reaches immediately the equilibrium with drying air and mass transfer is completely controlled by internal diffusion (D Model). This boundary condition is shown in Eq. 2 for the x mass transfer direction and the model's solution, in terms of the average moisture content, is illustrated in Eq. 3 (Crank, 1975; Simal et al., 1998).

$$W_p(L,y,z,t>0) = W_e \quad \text{(Eq. 2)}$$

$$W(t) = W_e + (W_c - W_e)\left[\sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 \pi^2} \exp\left(-\frac{D_e (2n+1)^2 \pi^2 t}{4L^2}\right)\right]^3 \quad \text{(Eq. 3)}$$

Where W is the particle average moisture content (kg water/kg dm), L the half length of cubic side (m) and subscripts c and e refer to critical and equilibrium states, respectively. Sorption data at -10 ºC reported by Claussen et al. (2007) were used to estimate equilibrium moisture content.

The D Model was fitted to the experimental data in order to identify the effective moisture diffusivity (D_e). The objective function to be minimized was the sum of the squared difference between experimental (W_{exp}) and calculated (W_{calc}) average moisture content. The optimization was conducted by applying the Generalized Reduced Gradient available in Solver tool (Microsoft Excel2007^{TM}, Microsoft, USA)

As a second approach, boundary layer thickness was not considered negligible and mass transfer is jointly controlled by diffusion and convection (D+C Model). Eq. 4 reflects this boundary condition for the x direction, representing the equality of diffusion and convection water fluxes in the interface. The D+C Model allows the quantification of both the effective diffusivity and the external mass transfer coefficient (k, kg water/m s).

$$t > 0 \qquad x = L \qquad -D_e \rho_{ds} \frac{\partial W_p(L,y,z,t)}{\partial x} = k\left(a_w(L,y,z,t) - \varphi_{air}\right) \qquad \text{(Eq. 4)}$$

Where ρ_{ds} is the dry solid density (kg dm/m^3), k the mass transfer coefficient (kg water/m^{-2}s), and φ_{air} the relative humidity of drying air. As aforementioned, the water activity in the surface of the material ($a_w(L,y,z,t)$) was estimated from sorption isotherms data reported in the literature (Claussen et al., 2007).

The D+C Model was solved by an implicit finite difference numerical method (Garcia-Perez et al., 2011) creating a computational algorithm in Matlab® 7.9.0 (The MathWorks, Inc., USA). The program provided the local moisture distribution inside the solid and the average moisture content of the solid, both as function of the drying time, the effective moisture diffusivity (D_e) and the mass transfer coefficient (k). As in the D model, kinetic parameters (k and D_e) were jointly identified by fitting the model to the experimental drying kinetics. The same objective function was used than in the D model, but in this case, the SIMPLEX method (fminsearch function) available in Matlab was used for optimization.

Both D and D+C Models were fitted to each drying kinetic replicate and kinetic parameters averaged. The analysis of variance (ANOVA) was carried out and LSD intervals ($p<0.01$) were estimated using Statgraphics® Plus 5.1 (StatPoint, Inc., USA) to evaluate the significant influence of ultrasound on D_e and k parameters. Finally, the explained variance (VAR, Eq. 5) was calculated in order to determine the goodness of the fit to the experimental data.

$$VAR=\left[1-\frac{S_{tw}^2}{S_w^2}\right]\times 100 \qquad \text{(Eq. 5)}$$

Where S^2_W and S^2_{tW} are the variance of the sample and the estimation, respectively.

3. Results and discussion

3.1. Drying kinetics

Figure 2 shows the AIR and AIR+US drying kinetics of carrot, apple and eggplant carried out at a temperature (-14 ºC) below product's freezing point. The low temperature keeps vapor pressure below the triple point and water removal mainly happens by sublimation. In such a way, this kind of experiments is commonly referred to as atmospheric or convective freeze drying.

Initial moisture contents were 7.58±0.90, 6.10±0.37 and 14.57±0.27 kg water/kg dry matter for carrot, apple and eggplant, respectively. The natural variability of materials being dried is showed by the drying curves in Figure 2, where it is observed that eggplant was the most heterogeneous material. Drying kinetics did not exhibit a constant rate period, which has been also reported for other products dried at low temperature (Wolff and Gibert, 1990 a and b; Kudra and Mujumdar, 2009). As a consequence, initial moisture content was considered the critical one for modeling purposes. Due to the low temperature used, drying times in AIR experiments are longer than 100 hours for apple and carrot (low and medium porosity products) and around 25-30 hours for eggplant (high porosity product). The more open structure of eggplant facilitates the water vapor leaving the solid matrix by molecular diffusion, involving higher drying rates than carrot or apple. The influence of internal structure on drying kinetics at temperature below freezing point has been reported in the previous literature (Claussen et al., 2007).

The application of power ultrasound greatly sped-up the drying kinetics. AIR+US experiments involved average reduction of drying time between 65 and 70 % for all the products. Thus in carrot experiments, to reach a moisture content of 1 kg water/kg dm, AIR experiments were extended until approximately 110 h, while AIR+US experiments only until 35 h (time reduction 68 %). In the case of eggplant, to reach a moisture content of 2 kg water/kg dm, the application of power ultrasound reduced drying time from 20 to 7 hours (time reduction 70). These results point to the fact that the ultrasonic effect was not dependent on product structure, which results opposite to the behavior observed in experiments carried out at temperatures above the product freezing point. Thus, in previous tests conducted at 40 ºC, 1 m/s and applying higher acoustic power (37 against 19.5 kW/m^3) than in this work, drying time was shortened by 32 % for carrot (Garcia-Perez et al., 2009) and 72 % for eggplant (Garcia-Perez et al., 2011). The fact that carrot and eggplant became different was related to porosity, thus large intercellular spaces of high porosity products make the product more prone to alternating compression and expansion cycles produced by ultrasonic waves (sponge effect) (Garcia-Perez et al., 2009). In the experiments carried out at 40 ºC, shrinkage is a significant phenomenon, which keeps the product’s porosity almost constant during drying due to water loss is buffered by sample volume reduction. Nevertheless, when drying at -14 ºC, the shrinkage is small and regardless the initial structure, all the products are converted in high porosity matrixes. During drying at low temperature, and considering the most commonly adopted mechanistic theory of the “uniformly ice retracting front” (Claussen et al., 2007b; Wolf and Gilbert, 1990b), vapor diffusion happens in the dry outer layer, which is getting thicker as drying progresses and ice core remains in the inner part. This is the reason because of the influence of power ultrasound during drying at -14 ºC was similar for carrot, apple and eggplant since all of them could be considered high porosity dry materials.

First attempts of using sound waves to intensify drying process at low temperature date from 70's. Moy and DiMarco (1972 and 1970) tested both the ultrasonic application at vacuum and non-vacuum freeze drying. In the case of vacuum freeze drying (Moy and DiMarco, 1972), a direct coupling between the ultrasonic transducer and the sample was adopted and the results obtained showed increments of drying rate from negligible to 6 % in beef samples. The authors concluded that the meaningful results were related to the low efficiency of the ultrasonic system used, although highlighted the potential of this technology and pointed to a better development of the ultrasonic device in order to increase efficiency. In the case of non-vacuum freeze drying, Moy and DiMarco (1970) used a stem-jet whistle working at frequencies laid between 10.8 and 12.2 kHz, which are within the human hearing range. Experiments were conducted with distilled water and coffee and tea extracts and temperatures ranging -15 and -26 ºC and reported average increases of drying rate between 10 and 100 %. The main concern of this study is also related to the complex ultrasonic application in gas media, the low frequency used partially avoids the acoustic energy attenuation; this action however, may involve an intense noise that could be an obstacle to its use in industrial applications. More efforts in this field have been recently done by Bantle and Eikevik (2011) using a commercial transducer (Sonotronic, DN 20/2000, 20 kHz) and testing the influence of process variables, such as temperature and power applied. These authors showed a maximum reduction of drying time by ultrasonic application around 10 % for drying green peas at -3 ºC. This improvement is almost negligible compared to the results obtained in this work, and could be related to the own variability of material being dried (see Figure 2). Although the experimental reproducibility was within ± 2 %, the authors did not clarify if the improvement was significant since the statistical study to validate the results consistency is missed. In addition, as pointed out by Bantle and Eikevik (2011), the results obtained were limited by the low efficiency of the ultrasonic application system used.

Figure 3 shows the ethanol removal kinetics from AIR dry apple, which were carried out with the only aim of testing the ability of ultrasound to remove other solvent different than water from a solid matrix. In this case, the removal occurs by evaporation due to the freezing point of ethanol (96 % v/v) is below -14 ºC. Ultrasound application also involved the increase of the drying rate, thus, the average time needed to remove completely the ethanol was reduced from 150 minutes (AIR) to 67.5 (AIR+US). This means a reduction of process time of approximately 120 %. These results open a potential application of this technology in chemical, cosmetic and pharmaceutical industry to remove organic solvents at low temperature in order to preserve quality aspects with lower processing costs than freeze drying. One of the main concerns in the use of organic solvents is the removal of traces. The cyclic and repeated vibration in the particle brought about by the ultrasonic waves could positively contribute to remove the organic solvent traces. Obviously, this hypothesis is still a challenge due to it has to be tested using more accurate detection methods than in this work, where ethanol content was monitored by weighting (±0.01 g) the samples at preset times.

3.2. Modeling drying kinetics

Modeling should concisely contribute to quantify and gain insight into the influence of ultrasound on mass transfer mechanisms involved during drying at low temperature (below freezing point). As already mentioned, the most commonly adopted mechanistic theory to describe how water removal occurs during atmospheric freeze drying is the “uniform ice retracting front” (Claussen et al., 2007b; Liapis and Bruttini, XXXX). This model considers that during drying the product is divided into two layers: a frozen inner core and a dry outer layer, being assumed that frozen core gradually and uniformly shrinking down to cero. Sublimation occurs in the ice front and water vapor moves through the dry layer to the sample surface. Therefore, mass transfer may be controlled by the internal vapor diffusion or by the external convection. According to literature (Bantle and Eikevik, 2011), water is primarily controlled by internal diffusion during the atmospheric freeze drying, thus modeling may be based predominantly on diffusion. Actually, comprehensive approaches based on the “uniform ice retracting front” theory need an accurate knowledge of thermal product properties, vapor diffusion coefficients and specific porosity (Heldman and Hohner, 1974), which are temperature dependent and change during drying. Therefore, multiple assumptions and hypothesis have to be done in order to simplify the model and, sometimes, the mechanistic goal is missed and modeling approaches to empiricism. In addition, the experimental validation of the “uniform ice retracting front” models results very complicated in foodstuffs and is still a challenge. Due to modeling was not the final goal of this work, general diffusion models were used, which commonly have a good behavior for air drying at temperatures below freezing point.

As a first approach, drying kinetics were modeled considering a pure diffusion model (D Model) neglecting the influence on drying rate of external air flow. The effective moisture diffusivity identified from experimental kinetics, as well the explained variance attained, is included in Table 1. In AIR experiments, D_e values were 1.1×10^{-11}, 1.6×10^{-11} and 4.8×10^{-11} m^2/s for carrot, apple and eggplant, respectively. The higher the initial product porosity, the higher the effective moisture diffusivity was found. Although, vapor diffusion occurs in the dry product layer, which is in all cases a high porosity material due to shrinkage being small, there still exists some differences in the tortuosity of water pathway due to the initial raw material properties. Due to the temperature used, D_e figures were one order of magnitude lower that the ones identified at temperatures around 30-40 ºC for conventional hot air drying experiments (Garcia-Perez et al., 2009 and 2011). Furthermore, it should be remarked that the figures identified in this work would be overcome considering the “uniform ice retracting front” theory due to diffusion distance was considered as constant (L) and not variable, from 0 to L, as function of drying time or moisture content. AIR drying kinetics of carrot showed a clear diffusion behavior, being the average VAR provided by the D Model of 99.3 %. This fact is also observed in Figure 4, where the fit of the diffusion model to the experimental data is shown. As the initial product porosity and consequently the effective diffusivity increased, the external resistance started to be a significant phenomenon on the mass transfer control. Thus, for apple drying without ultrasound application, the drying kinetic was almost completely controlled by diffusion (VAR 98.0 % from Model D), such as is observed in Figure 4. However, in the case of eggplant drying, the VAR attained with the D Model was low (93.4 %), which suggests that the influence of the external mass transfer should be considered.

The application of power ultrasound increased the effective diffusivity, the increase on this parameter ranged from 182 % to 244 % for carrot and apple, respectively. In all the products, the increase of the effective moisture diffusivity was significant at a significance level of 99 %. The improvement was really evident and states the high efficiency of the ultrasonic system used in this work. Bantle and Eikevik (2011) reported an increase of the effective diffusivity of up to 14.8 % in drying of green peas at -6 ºC by power ultrasound application. The explained variance attained through model D in the AIR+US experiments was lower than in the AIR ones for all the products. This fact means an influence of ultrasound on mass transfer control mechanism. Thus in carrot drying, the VAR changed from 99.3 to 91.8 %, which highlights that diffusion was not the only significant mechanism controlling water removal and suggests an influence of external resistance to mass transfer when ultrasound is applied. Therefore, it seems that ultrasound had a different effectiveness over internal and external mass transfer mechanisms. In order to clarify this issue, a diffusion model including external resistance to mass transfer was considered (D+C Model).

The results of D+C Model are shown in Table 1, the explained variance was higher than 99.5 % in all cases, which states the significant influence of external resistance on experimental drying kinetics. The goodness of the fit achieved with the D+C Model is shown in Figure 5 for eggplant. In AIR eggplant drying and comparing with the D Model, VAR increased from 93.4 to 99.9 % (Figure 5) when considering external resistance (model D+C), being the observed increments lower for apple and carrot AIR experiments where mass transfer was mainly controlled by internal diffusion. In AIR+US drying experiments, VAR ranged from 99.8 to 99.9%, being much higher than the figures attained with the D Model (from 91.8 to 92.3) (Table 1). Therefore, as already mentioned, when ultrasound was applied the importance of internal diffusion on mass transfer control was reduced. This fact is explained from the relative increase of effective moisture diffusivity and mass transfer coefficient. Average improvements from 96 to 170 % were found for the mass transfer coefficient when AIR and AIR+US experiments are compared. However, the increase of the effective moisture diffusivity was laid between 407 and 428 %, which indicates that the relative importance of internal diffusion over mass transfer control was reduced when ultrasound was applied and convection plays a key role. The differences for D_e and k between AIR and AIR+US experiments were significant at a confidence level of 99 %. Maximum increases for D_e and k around 100 % have been reported in previous works carried at temperatures of 30-40 ºC with different vegetables applying acoustic powers higher than in this work (García-Perez et al., 2006; Garcia-Perez et al., 2011 b and Ozuna et al., 2011). The alternating compression and expansion cycles produced by ultrasonic waves (sponge effect) should facilitate the vapor diffusion through the solid matrix. In such a way, the ultrasonic effect should not be located in the ice core if not in the dry outer layer in which vapor diffusion occurs. This is why the ultrasonic effect on the effective diffusivity was quite similar for all products (Table 1). Due to the high porosity of the dry layer, the effects brought about by compression and expansion cycles produced by ultrasonic waves are more intense than in conventionally hot air dried products where the small and water filled intercellular spaces did not allow such behaviour and diminish the ultrasonic effects (Garcia-Perez et al., 2010; Garcia-Perez et al, 2009). The high efficiency of ultrasound application over diffusion should be also linked to the greater acoustic energy absorption in high porosity products (Garcia-Perez et al., 2010). Finally, the ultrasonic effects on the external mass transfer resistance should be similar to those reported for conventional hot air drying

(Muralidhara et al., 1985; Gallego-Juarez et al., 1999). Pressure variations, oscillating velocities and microstreaming affect the solid-gas interfaces reducing boundary layer thickness and, therefore, improving the water transfer rate from the solid surface to the air medium.

4. Conclusions

In this work, the feasibility of power ultrasound application to intensify the mass transfer rate on air drying at low temperatures (below freezing point) has been illustrated. Power ultrasound application led to a drastic reduction of drying times (around 65-70 %) due to the improvement of both internal vapor diffusion and external convection. Thus, effective moisture diffusivity and mass transfer coefficient increased from 96 to 170 % and from 407 and 428 %, respectively. These results showed the high efficiency of the ultrasonic application system used compared to previous literature. In addition, the ability of ultrasound to speed up the removal of ethanol from a solid matrix has been also evidenced, which opens a future application field of this technology not only in food but also in chemical, pharmaceutical and cosmetic industries to remove organic solvents preserving product quality attributes.

Ultrasound application should be considered a potential and effective technology to intensify low temperature drying processes, being capable to make more affordable and less energy and time-consuming these processes for all kind of industries. Future studies should be addressed to determine the influence of process parameters (air velocity and temperature, acoustic power or mass density) and optimize the ultrasonic application aiming to minimize energy consumption. Comprehensive heat and mass transfer mechanistic models considering the “uniformly ice retracting front” theory should be developed, solved, evaluated and validated. In addition, more efforts will be carried out in the design and development of new ultrasonic devices to be more efficient. Although, all these future works will have as primary goal the development of ultrasound technology for industrial applications.

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FIGURE CAPTIONS

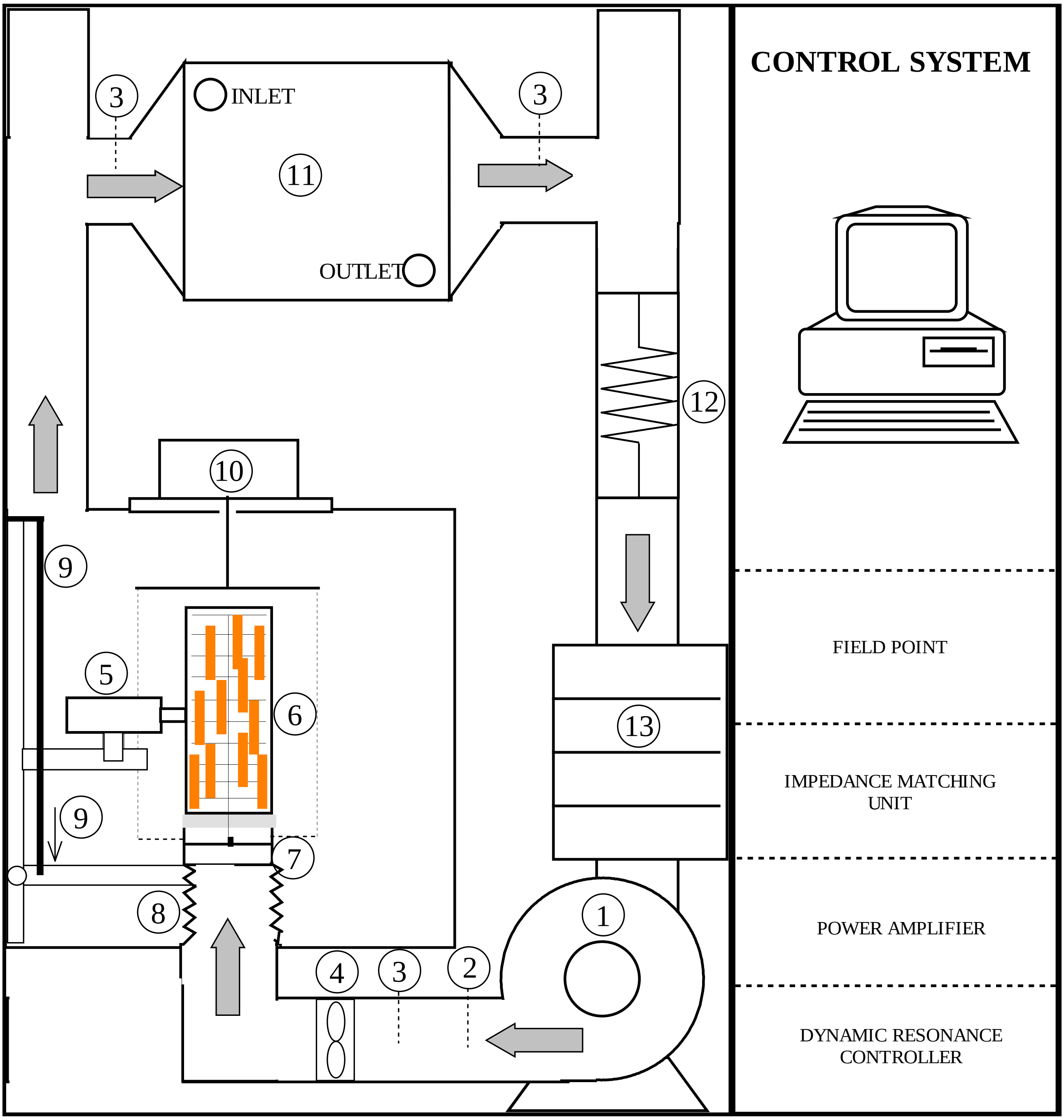
Figure 1. Diagram of the ultrasonically assisted convective drier. 1. Fan, 2. Anemometer, 3. Temperature and relative humidity sensor, 4. 3-Way valve, 5. Ultrasonic transducer, 6. Vibrating cylinder, 7. Sample load device, 8. Retracting pipe, 9. Slide actuator, 10. Weighing module, 11. Heat exchanger, 12. Heating elements, 13. Desiccant tray chamber. 14. Pt-100.

Figure 2. Experimental drying kinetics of carrot, apple and eggplant. AIR: Conventional drying experiments (-14 ºC, 1 m/s) and AIR+US: Ultrasonically assisted drying experiments (-14 ºC, 1 m/s, 19.5 kW/m^3).

Figure 3. Kinetics of ethanol removal from AIR dried apple. AIR: Conventional drying experiments (-14 ºC, 1 m/s) and AIR+US: Ultrasonically assisted drying experiments (-14 ºC, 1 m/s, 19.5 kW/m^3).

Figure 4. Fit of the D model to the drying kinetics of carrot and apple. AIR: Conventional drying experiments (-14 ºC, 1 m/s) and AIR+US: Ultrasonically assisted drying experiments (-14 ºC, 1 m/s, 19.5 kW/m^3). In each plot, only one replicate is included.

Figure 5. Fit of the D+C model to the drying kinetics of eggplant. AIR: Conventional drying experiments (-14 ºC, 1 m/s) and AIR+US: Ultrasonically assisted drying experiments (-14 ºC, 1 m/s, 19.5 kW/m^3). In each plot, all the replicates are included with the simulation.



573
574
575
576 **Figure 1**
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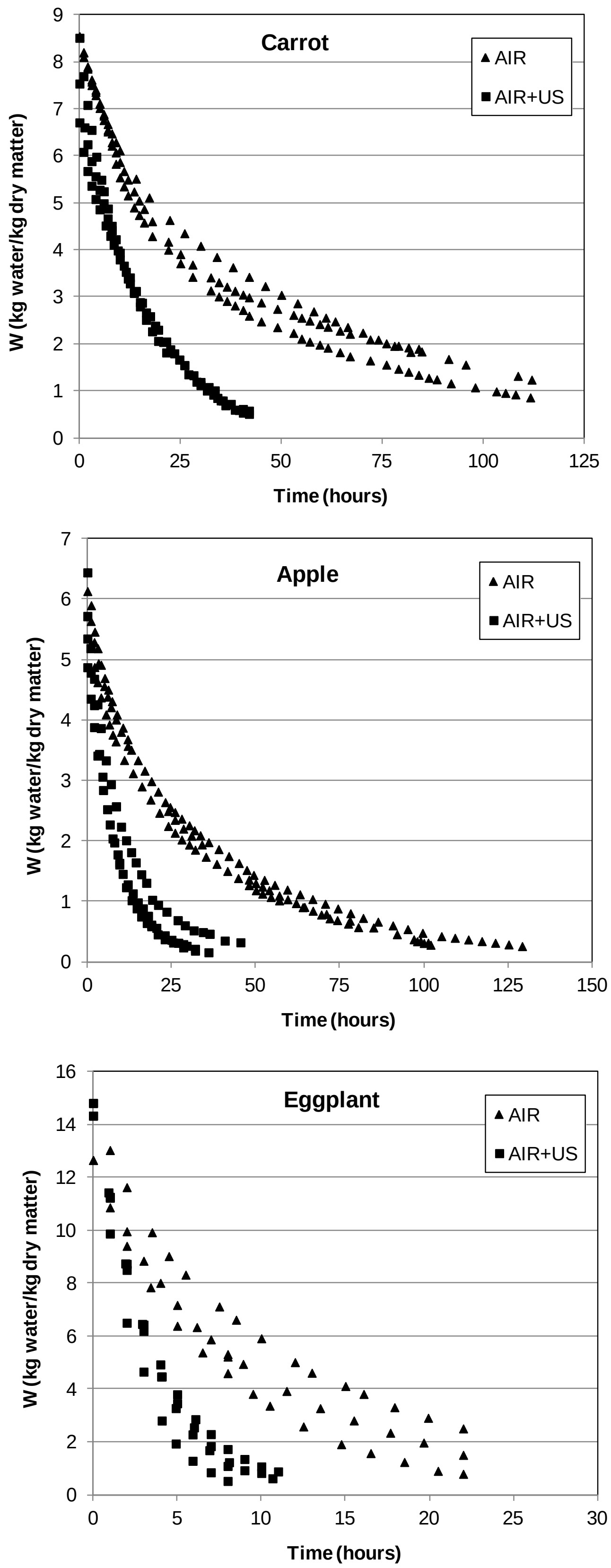
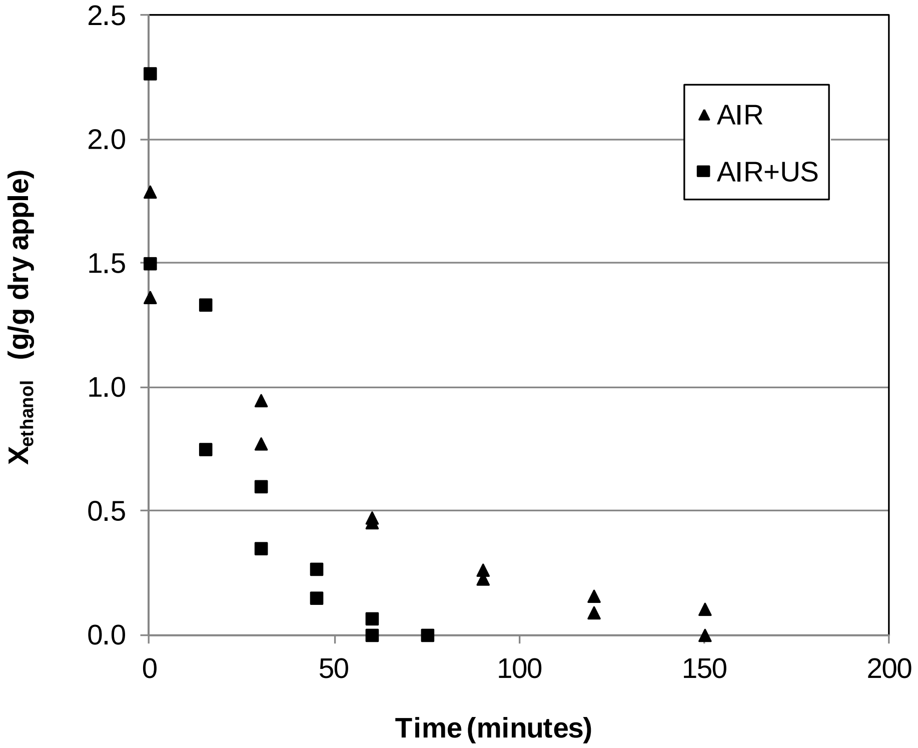

Carrot
AIR
AIR+US
W (kg water/kg dry matter)
Time (hours)
0
1
2
3
4
5
6
7
8
9
0
25
50
75
100
125
Apple
AIR
AIR+US
W (kg water/kg dry matter)
Time (hours)
0
1
2
3
4
5
6
7
0
25
50
75
100
125
150
Eggplant
AIR
AIR+US
W (kg water/kg dry matter)
Time (hours)
0
2
4
6
8
10
12
14
16
0
5
10
15
20
25
30

Figure 2



2.5
2.0
1.5
1.0
0.5
0.0
0
50
100
150
200
AIR
AIR+US
$X_{ethanol}$ (g/g dry apple)
Time (minutes)

Figure 3

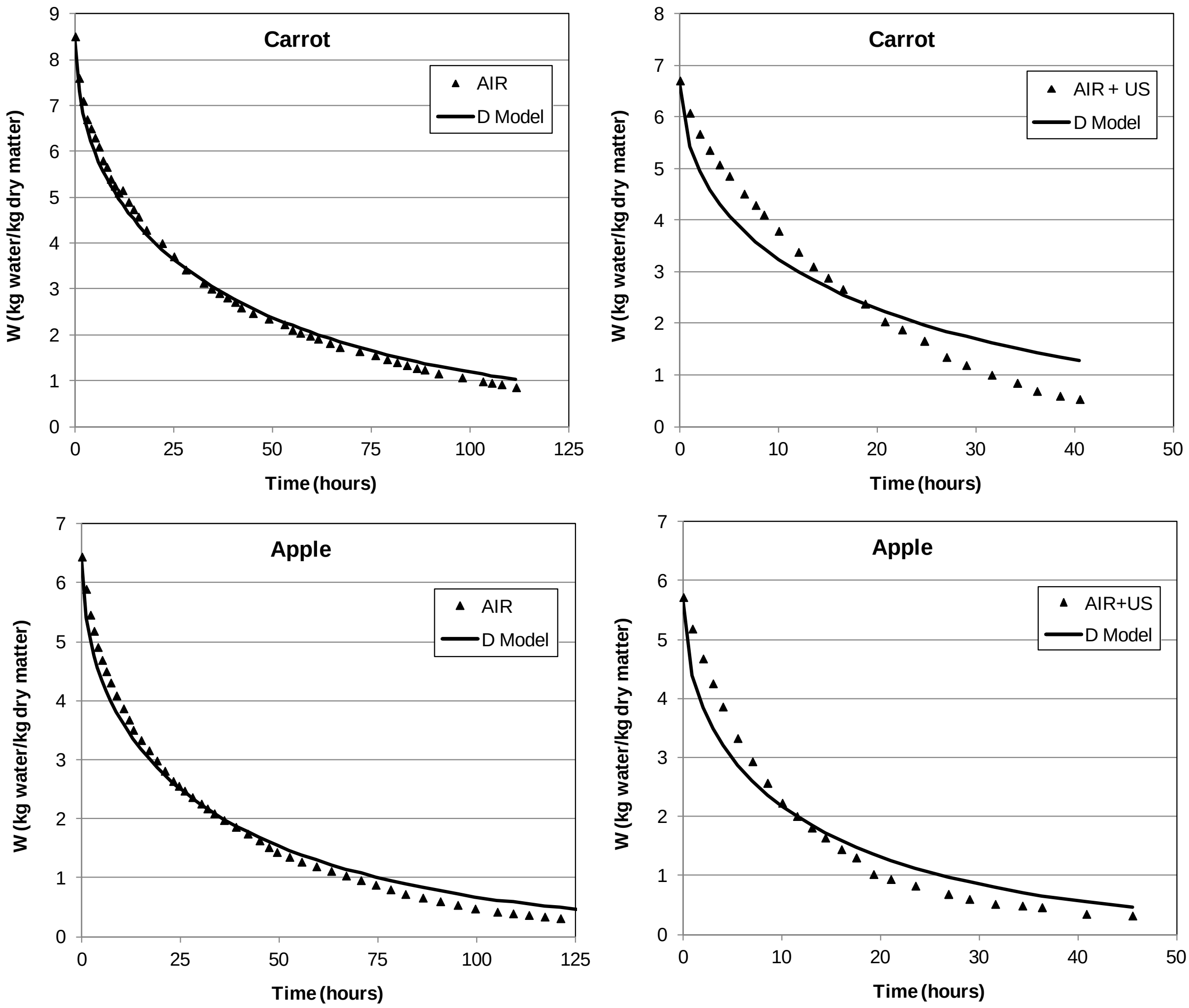

Carrot
AIR
D Model
W (kg water/kg dry matter)
Time (hours)
Carrot
AIR + US
D Model
W (kg water/kg dry matter)
Time (hours)
Apple
AIR
D Model
W (kg water/kg dry matter)
Time (hours)
Apple
AIR+US
D Model
W (kg water/kg dry matter)
Time (hours)

Figure 4

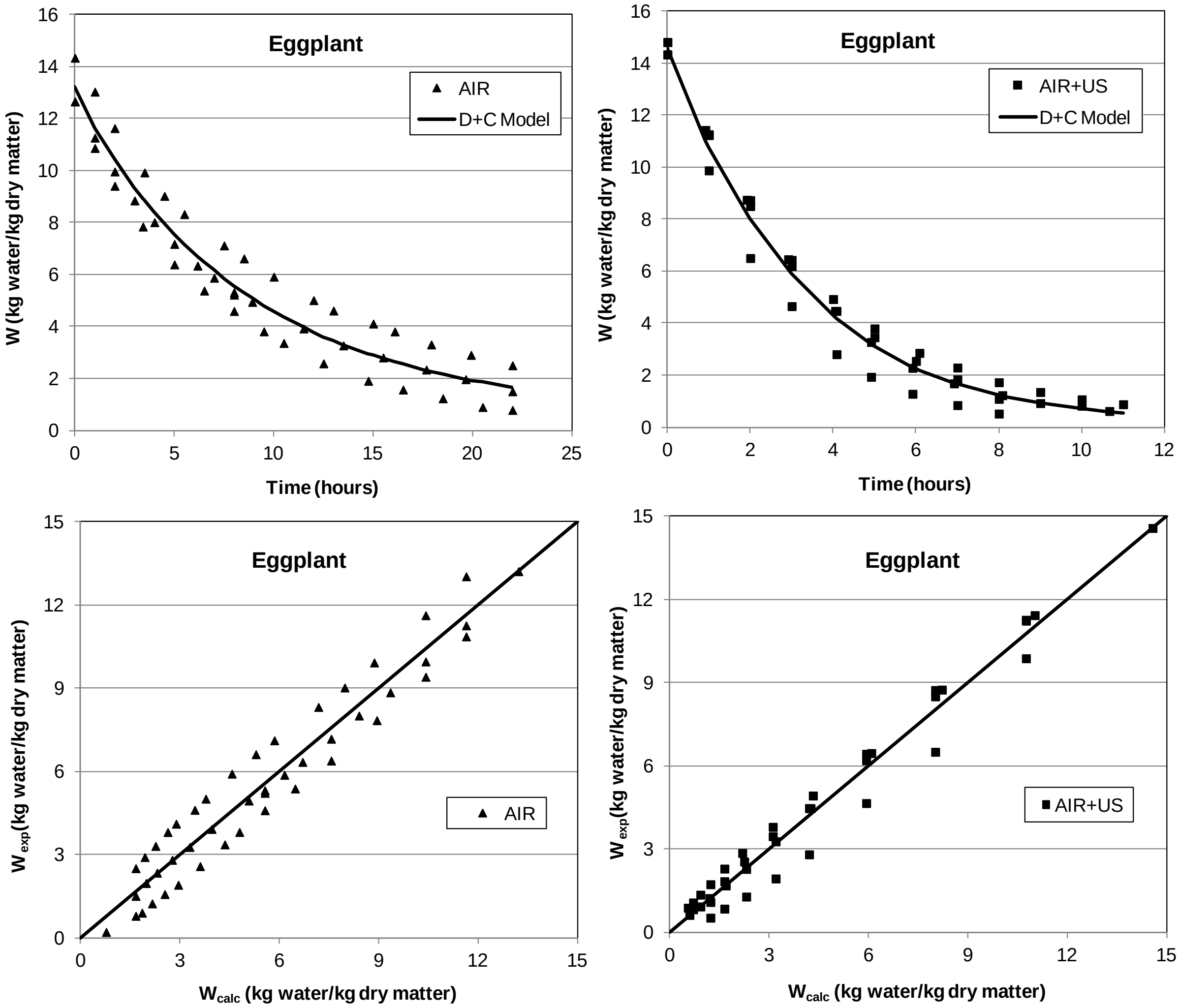

Eggplant
AIR
D+C Model
W (kg water/kg dry matter)
Time (hours)
Eggplant
AIR+US
D+C Model
W (kg water/kg dry matter)
Time (hours)
Eggplant
AIR
W_{exp}(kg water/kg dry matter)
W_{calc} (kg water/kg dry matter)
Eggplant
AIR+US
W_{exp}(kg water/kg dry matter)
W_{calc} (kg water/kg dry matter)

Figure 5

Table 1. Results of drying kinetics modeling. Average values and standard deviation of kinetic parameters identified from D and D+C Models. Increment shows (in percentage) the increase of a kinetic parameter by the application of ultrasound.

		D MODEL		D+C MODEL		
		D_e (10^{-11} m^2/s)	**VAR (%)**	**D_e (10^{-11} m^2/s)**	**k (10^{-5} kg water/m^2 s)**	**VAR (%)**
Carrot	**AIR**	1.1±0.1	99.3	0.8±0.1	3.3±1.5	99.6
	AIR+US	3.1±0.3	91.8	4.2±0.4	8.3±2.3	99.8
	Increment (%)	182		425	152	
Apple	**AIR**	1.6±0.4	98.0	1.4±0.7	4.8±0.2	99.5
	AIR+US	5.5±1.1	93.3	7.4±2.1	9.4±0.9	99.9
	Increment (%)	244		428	96	
Eggplant	**AIR**	4.8±1.3	93.4	4.4±1.7	23.7±4.3	99.9
	AIR+US	15.8±3.3	92.3	22.3±4.7	64.1±10.4	99.9
	Increment (%)	229		407	170	