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Drying intensification combining ultrasound pre-treatment and ultrasound-assisted air drying

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ABSTRACT

The water effective diffusivity can be increased in air drying of fruits and vegetables by applying an ultrasonic pre-treatment in ultrasonic baths or by directly applying ultrasound during air-drying. This work has examined the influence of the combination of both techniques on the dehydration of apple cubes (*Malus domestica* L. var Royal Gala). Experiments were carried out drying apples using conventional convective air-drying, ultrasound-assisted air-drying, ultrasound pre-treatment followed by conventional air-drying and ultrasound pre-treatment followed by ultrasound-assisted air-drying. The drying kinetics was modeled assuming internal and external resistances to mass transfer. The results showed that the effects of both ultrasonic processes are additive and that the use of the combined technique can reduce air drying time. The combination of the ultrasonic pre-treatment and ultrasound-assisted air-drying has intensified the drying process imparting in an increase of the water effective diffusivity by up to 93%, an increase of the external mass transfer by up to 30% and a reduction of up to 58% in the total drying time when compared to the conventional air-drying process.

1. Introduction

The use of ultrasound in drying of fruits has been carried out in two ways: using ultrasound pre-treatments prior to conventional drying and using air-borne ultrasonic drying (Cárcel et al., 2012; Fernandes and Rodrigues, 2008a; Mulet et al., 2003). The two techniques have proven to reduce the total drying time and to increase the water effective diffusivity during air-drying (Dehghannya et al., 2016; Fijalkowska et al., 2016; Schossler et al., 2012). In this work, we combine the two techniques to evaluate if there is a synergistic effect on drying when both techniques are used.

Ultrasound pre-treatment has been applied to several fruits, such as apples, pineapples, sapotas, melons, genipap, papaya and Malay apples (Fernandes and Rodrigues, 2012, 2008b; Fernandes et al., 2007; Garcia-Noguera et al., 2010; Mothibe et al., 2014; Oliveira et al., 2010; Rodrigues and Fernandes, 2007). In most of these fruits, the ultrasound pre-treatment has increased the water effective diffusivity leading to a reduction in the total drying time between 10 and 50%. The increase in the water effective diffusivity was attributed to the formation of microscopic channels during ultrasound pre-treatment (Fernandes et al., 2009, 2008a; Rodrigues et al., 2009).

Air-borne ultrasound has also proven to increase the water effective diffusivity and to reduce drying time (Beck et al., 2014; Schossler et al., 2012). The technique has been applied to apples, potato, carrots, orange peel, tomato and eggplant, and showed to increase the water effective diffusivity between 40 and 70% (Fernandes et al., 2016; García-Pérez et al., 2006, 2012; Ozuna et al., 2011). Water removal is influenced by both internal and external resistances. Air-borne ultrasound can be effective in reducing the external mass transfer resistance, which is influenced by airspeed, air direction, humidity, temperature and the interface between the fruit and air phase (Beck et al., 2014).

The combined technique applying the ultrasonic pre-treatment followed by ultrasound-assisted air-drying is the focus of this study. To evaluate the synergistic effect of both ultrasound techniques,

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experiments were carried out drying apples using conventional convective air-drying, ultrasound-assisted air-drying, ultrasound pre-treatment followed by conventional air-drying and ultrasound pre-treatment followed by ultrasound-assisted air-drying, under different drying conditions (temperature and air velocity). The discussion of the synergistic effect was carried out analyzing the internal and external resistances to mass transfer through a kinetic model, and by means of analyzing the tissue structure of the dried samples.

2. Materials and methods

2.1. Preparation of samples

Apples (*Malus domestica* L. var Royal Gala) were bought from the local market (Valencia, Spain). Only fruits with same maturity stage were used. Apples were cut into cubes of 8.5 × 8.5 × 8.5 mm. The moisture content of the fruit was determined by heating in a drying oven at 60 °C for 24 h under vacuum (50 mmHg).

2.2. Ultrasonic pre-treatment

The ultrasonic pre-treatment was carried out with a probe ultrasonic system (Hielscher model UP 400s) using a 13 mm probe and 160 W of power. The ultrasonic power was measured using the calorimetric method (Löning et al., 2002).

An amount of 40 g of apples was immersed in 400 mL of distiller water and subjected to ultrasound at a power density of 400 W/L for 10 min. This sonication time allows the formation of microscopic channels in fruit samples as showed in previous studies of our group and other researchers (Fernandes and Rodrigues, 2008a; Garcia-Noguera et al., 2010). After the pre-treatment the samples were blotted with paper tissue to remove water in excess and were transfer to the air-drier. All experiments were carried in triplicates.

The moisture content of the apples before and after the pre-treatment were measured using 15 g of apples divided into three 5 g assays to have triplicates of the moisture content. The moisture content of the fruit was determined by heating in a drying oven at 60 °C for 24 h under vacuum (50 mmHg).

2.3. Air drying

Drying kinetics was carried out in an ultrasound-assisted convective drier (Cárcel et al., 2007; García-Pérez et al., 2009; Ortuño et al., 2010). In this dryer, the drying chamber is comprised by an aluminum-vibrating cylinder (internal diameter 10 cm, height 31 cm, and thickness 1 cm) driven by a piezoelectric composite transducer (21.7 kHz) that produces the acoustic waves.

Air-drying experiments of apple samples were carried out at three different air velocities (1, 2 and 3 m/s) and two different temperatures (45 and 60 °C). Drying experiments assisted by ultrasound were carried out under the same experimental conditions as air-drying experiments and by applying an electric power of 75 W to the ultrasound transducer. The drying experiments were conducted in duplicate and completed when samples lost 80% of the initial weight. Drying kinetic was determined from the initial moisture content and the weight loss logged during drying. For each experiment, 40 cubes of apple were placed in a custom sample holder inside the drying chamber, weighing 25 g of fruit.

2.4. Modeling

The air-drying kinetics was modeled considering both internal and external mass resistances. Only the falling-rate period (diffusion-controlled mass transfer period) was considered because during the experiments the constant-rate period (heat transfer-controlled mass transfer period) was not observed.

The finite volume method was used to model the kinetics and water content inside the apple cubes. Mass balance inside the control volume was taken as the second Fick Law (Equation (1)).

$$\frac{\partial W}{\partial t} = D\left(\frac{\partial^2 W}{\partial x^2} + \frac{\partial^2 W}{\partial y^2} + \frac{\partial^2 W}{\partial z^2}\right) \tag{1}$$

Equation (1) was discretized using the progressive differences for the variable time and using the centered differences method for the space variables (x, y and z). The discretized form of Fick's second law is presented in Equation (2).

$$\begin{aligned} W_{t+1,i,j,k} = W_{t,i,j,k} + (D.\Delta t)\Bigg[&\left(\frac{W_{t,i+1,j,k} \; - \; 2W_{t,i,j,k} \; + \; W_{t,i-1,j,k}}{\Delta x^2}\right) \\ &+ \left(\frac{W_{t,ij+1,k} \; - \; 2W_{t,ij,k} \; + \; W_{t,i,j-1,k}}{\Delta y^2}\right) \\ &+ \left(\frac{W_{t,i,j,k+1} \; - \; 2W_{t,i,j,k} \; + \; W_{t,i,j,k-1}}{\Delta z^2}\right)\Bigg] \end{aligned} \tag{2}$$

Equation (2) was subjected to two contour conditions. Given the symmetry of the apple cubes, the center of the solid is the place where the moisture content assumes its maximum value, and thus, the derivatives of the moisture content regarding the center planes are equal to zero (Equations (3)–(5)).

$$\frac{\partial W}{\partial z} = \frac{\partial^2 W}{\partial z^2} = 0 \tag{3}$$

$$\frac{\partial W}{\partial x} = \frac{\partial^2 W}{\partial x^2} = 0 \tag{4}$$

$$\frac{\partial W}{\partial y} = \frac{\partial^2 W}{\partial y^2} = 0 \tag{5}$$

The second contour condition was related to the flow of water from the solid phase (apple cube) to the air phase. For this contour condition, it was considered that the internal diffusivity flow rate was equal to the convective flow rate in the surface of the solid, representing the external resistance to mass transfer (Equations (6)–(8)).

$$\frac{\partial W}{\partial z} = \frac{Kc}{D\rho}(W_\infty - W_{sup}) \tag{6}$$

$$\frac{\partial W}{\partial x} = \frac{Kc}{D\rho}(W_\infty - W_{sup}) \tag{7}$$

$$\frac{\partial W}{\partial y} = \frac{Kc}{D\rho}(W_\infty - W_{sup}) \tag{8}$$

The Biot Number was calculated to evaluate the influence of the internal and external mass resistances during the drying process (Equation (9)).

$$Bi_M = \frac{Kc}{D}\left(\frac{V_s}{A_s}\right) \tag{9}$$

Given the symmetry of the apple cubes, the cube was divided in 8 corners and one corner was considered when implementing the solving algorithm. A mesh sensitivity evaluation was carried out for the finite differences method and it has indicated that 8 segments should be used in each direction. More segments increased the computer processing time and did not change the quality of the results (parameter estimation and fitting of the experimental results). As such, the solving algorithm was built considering the apple cube as being comprised of a total of 288 finite volumes.

The mean moisture content in the apple cube, at any given time, was obtained by numerical integration of the moisture content of the finite volume (Equation (10)).

$$\overline{W} = \frac{\int_0^{V_s/8} W dV}{\int_0^{V_s/8} dV} = 8\frac{\left(\sum_{k=0}^{a}\sum_{j=0}^{a}\sum_{i=0}^{a} W_{t,i,j,k}\ .V_c\right)}{V_s} \qquad (10)$$

To estimate the model parameters, D and K_C, an algorithm based on the Levenberg-Marquardt method was used. A computer program was written, using the language Python, to solve the model and to estimate the model parameters.

2.5. Tissue structure photomicrographs

The dried sample cubes were fixed with 4% solution of paraformaldehyde in 0.1 M phosphate buffer, pH 7.2 and 1% glutaraldehyde for 24 h at ambient temperature (Karnovisky, 1965). The material was dehydrated in a graded ethanol series and embedded in Historesin embedding kit (Jung). The tissue blocks were sectioned at 5 μm on a Leica RM 2065 microtome (Leica, Germany). The Periodic Acid-Schiff reagent (PAS) cytochemical reaction was employed for polysaccharide detection. Photomicrographs of the cell structure were taken using an Olympus BX51 light microscope (Olympus, Japan) with digital image capture system.

2.6. Statistical analysis

The results were evaluated by means of multifactorial ANOVA. The LSD (least significance difference) intervals ($p < 0.05$) were estimated. Statistical analysis was carried out using the software Statistica v.13 (Statsoft).

3. Results and discussion

The apple samples presented an initial moisture content of 83.9 ± 0.5 $g_{\text{water}}/100\ g_{\text{fresh fruit}}$ (wet basis) and an average density of 0.75 kg/cm^3. The drying kinetics of the apples samples presented the typical behavior observed for other fruits (Fernandes and Rodrigues, 2008a) and only the falling rate period was found. Dried samples presented a final moisture content of 0.30 ± 0.02 $g_{\text{water}}/g_{\text{dry matter}}$, which corresponded to a weight loss of 80% for the raw apple.

Fig. 1 shows the drying kinetics of the apples samples obtained with and without ultrasound pre-treatment and with and without ultrasound application during air-drying. The drying rates were higher for the process carried out with ultrasound pre-treatment + ultrasound-assisted air-drying followed by ultrasound-assisted air-drying, ultrasound pre-treatment + conventional air-drying, and conventional air-drying. The results show the synergism that can be obtained by associating the ultrasonic pre-treatment and the ultrasound-assisted air-drying.

The application of ultrasound reduced the time required for drying. A reduction up to 58% was observed comparing the results obtained for the combined ultrasound process and the conventional air-drying process (Table 1).

The reduction in drying time was also within the observed for other fruits (40–70%). Cárcel et al. (2007) found a 40% reduction for persimmon cylinders; Ortuño et al. (2010) found a 72% reduction

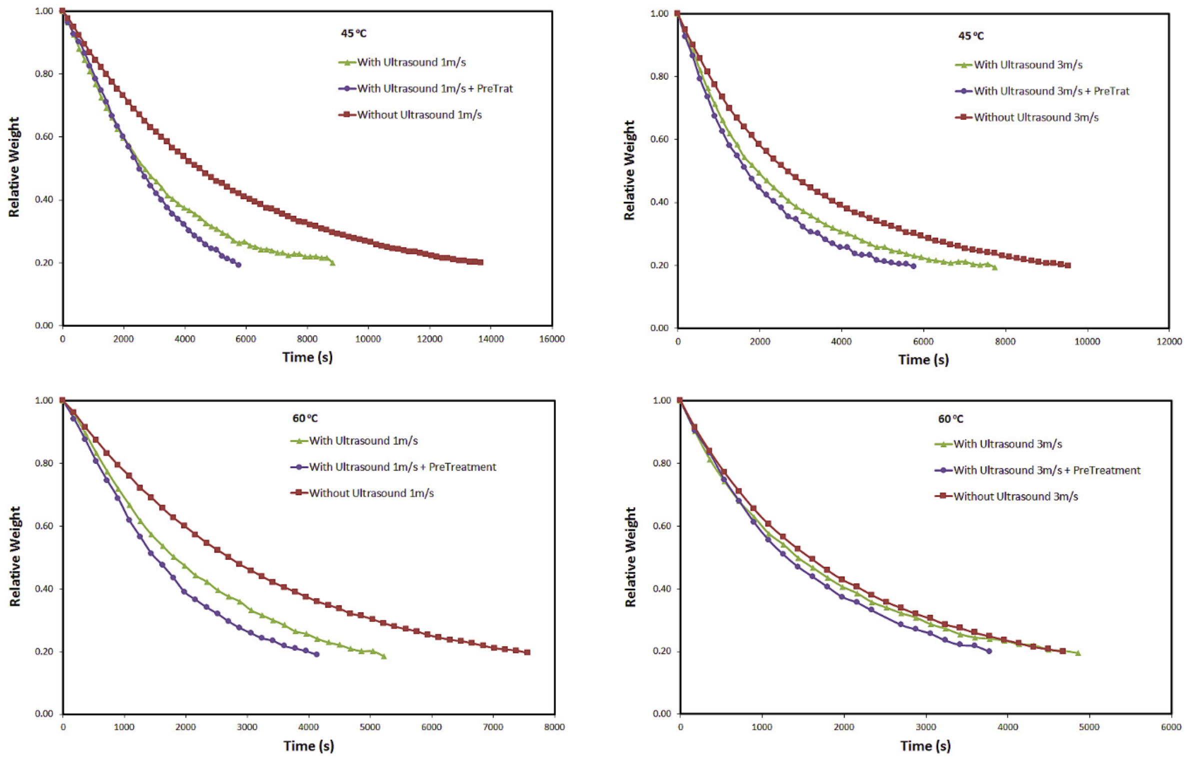

Fig. 1. Drying kinetics of apples cubes subjected to air-drying, ultrasound-assisted air-drying and ultrasound-assisted air-drying after ultrasonic pre-treatment. (a) 45 °C and air velocity of 1 m/s; (b) 45 °C and air velocity of 3 m/s; (c) 60 °C and air velocity of 1 m/s; (d) 60 °C and air velocity of 3 m/s.

Table 1
Reduction in drying time obtained by drying carried out with (USAir) and without ultrasonic application (Air), with or without subjected to ultrasound pre-treatment (PT) in comparison with conventional air-drying.

Temperature (°C)	Air velocity (m/s)	Time reduction PT + Air (min)	Time reduction USAir (min)	Time reduction PT + USAir (min)
45	1	39 (17%)	81 (35%)	132 (58%)
	2	42 (22%)	75 (40%)	90 (48%)
	3	21 (13%)	30 (13%)	63 (39%)
60	1	24 (19%)	39 (31%)	57 (45%)
	2	18 (17%)	15 (15%)	39 (38%)
	3	9 (11%)	0 (0%)	18 (22%)

for eggplant cylinders and 49% for orange peel slabs; and Ozuna et al. (2011) found a 40% reduction for potato cubes. The reduction in drying time observed for apples for the ultrasound-assisted air drying was also within the range observed for the process consisting of ultrasound pre-treatment followed by air-drying (10–50%). Fernandes, Linhares, & Rodrigues (2008), Fernandes & Rodrigues (2008a,b) and Rodrigues and Fernandes (2007) found reduction in the drying time of 10% for bananas and genipap slabs, 23% for sapotas cubes; 39% for melon cubes; 45% for pineapple cubes, while Oliveira et al. (2010) found a 28% reduction for Malay apples slabs.

The water effective diffusivities for apple drying and the external mass transfer coefficient in the different conditions tested herein are presented in Table 2. The diffusion model was adequate for describing the drying kinetics of apples cubes under the different experimental conditions tested. Chi-square test for the model showed levels of significance higher than 95% for all conditions, while R^2 values were higher than 0.97.

At 45 °C and 1 m/s of air velocity, the water diffusivity for the combined ultrasonic pre-treatment and the ultrasound-assisted process was 53% higher than the ultrasound assisted air-drying process and 93% higher than the conventional process. The external mass transfer coefficient was similar in the ultrasound assisted air-drying process with and without ultrasonic pre-treatment, and it was 59% higher than the conventional process.

At 60 °C and 3 m/s of air velocity, the water diffusivity for the combined ultrasonic process was 12% higher than the ultrasound assisted air-drying and 21% higher than the conventional air-drying. The external mass transfer coefficient, however decreased when the pre-treatment was applied and the ultrasound assisted air-drying process with ultrasonic pre-treatment was 15% lower than the same process without applying the ultrasonic pre-treatment. In both processes, the external mass transfer coefficient was higher than the value obtained in the conventional process (10% and 30% higher for the ultrasound-assisted air-drying process respectively with and without ultrasonic pre-treatment).

The values obtained for the water diffusivity and the external mass transfer coefficient show that the ultrasonic pre-treatment has a more significant effect on the internal water diffusivity, while the application of ultrasound during air-drying has a more significant effect on the external mass transfer coefficient. The combination of the ultrasonic pre-treatment with the application of ultrasound during air-drying showed a synergism increasing both internal and external mass transfers.

Although the airborne ultrasound showed a significant effect on increasing the external mass transfer, it has also increased the internal mass transfer. Ultrasound pressure waves seems to increase the moisture transport process improving the effective water diffusivity (Beck et al., 2014). The acoustic field was negatively affected by the air flow at high velocities and there was not a significant increase in the external mass transfer caused by the application of ultrasound under high air velocities.

The Biot number indicates that the process is mainly driven by internal resistance ($Bi_M > 600$), for all operating conditions that

Table 2
Effective water diffusivity, external mass transfer coefficients and Biot number for apple cubes dried at different temperatures and air velocities, carried out with and without ultrasonic application, with or without ultrasonic pre-treatment.

Temperature (°C)	Air velocity (m/s)	Effective Diffusivity (10^7 m^2/s)	External Mass Transfer (10^5 kg m^2/s)	Biot Number	R^2
Convective Air-Drying					
45	1	1.49 ± 0.10	3.19 ± 0.08	286.2	0.97
	2		4.46 ± 0.13	400.2	0.98
	3		5.58 ± 0.16	501.1	0.99
60	1	2.85 ± 0.22	5.32 ± 0.17	248.7	0.99
	2		7.09 ± 0.14	331.4	0.99
	3		9.91 ± 0.39	463.5	0.99
Ultrasonic Pre-Treatment followed by Convective Air-Drying					
45	1	3.60 ± 0.21	2.96 ± 0.08	109.7	0.98
	2		4.10 ± 0.12	151.8	0.98
	3		4.74 ± 0.19	175.5	0.99
60	1	6.46 ± 0.32	4.69 ± 0.11	96.9	0.98
	2		6.28 ± 0.16	129.7	0.99
	3		7.96 ± 0.25	164.4	0.99
Ultrasound-Assisted Convective Air-Drying					
45	1	3.75 ± 0.18	4.90 ± 0.11	174.3	0.97
	2		6.24 ± 0.15	221.9	0.98
	3		6.92 ± 0.30	246.1	0.99
60	1	5.09 ± 0.38	7.42 ± 0.20	194.4	0.98
	2		7.26 ± 0.18	190.1	0.99
	3		9.66 ± 0.31	253.0	0.99
Ultrasonic Pre-Treatment followed by Ultrasound-Assisted Convective Air-Drying					
45	1	5.03 ± 0.22	4.67 ± 0.10	123.8	0.99
	2		6.41 ± 0.15	169.9	0.99
	3		7.56 ± 0.22	200.6	0.99
60	1	7.36 ± 0.41	8.37 ± 0.22	151.7	0.99
	2		8.60 ± 0.25	155.9	0.99
	3		9.07 ± 0.24	164.3	0.99

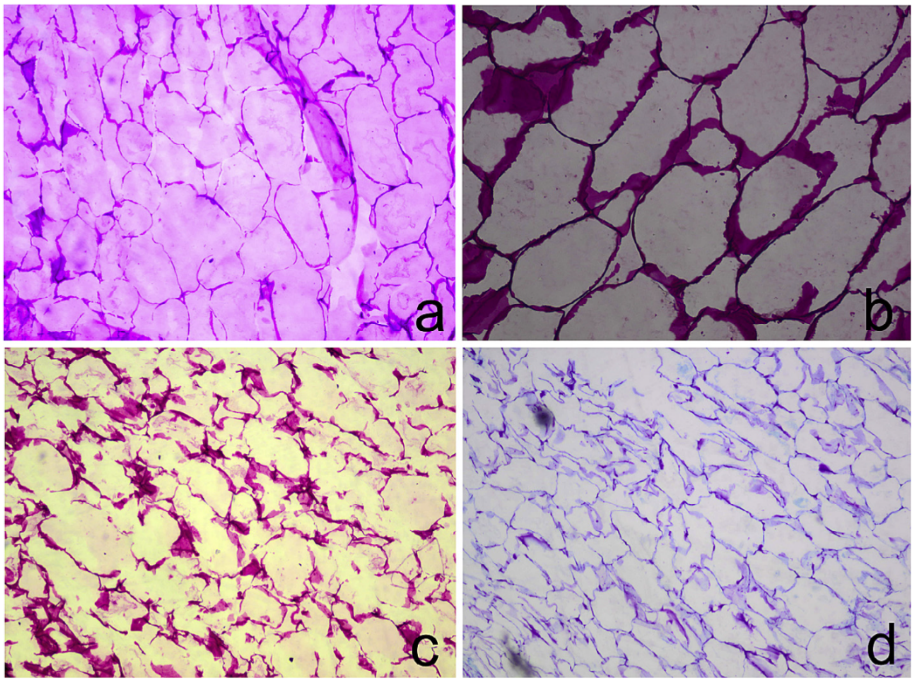

Fig. 2. Micrograph of apples tissue structure. (a) raw apple; (b) apples subjected to ultrasound pre-treatment; (c) apples subjected to ultrasound pre-treatment and air-dried; (d) apples subjected to ultrasound pre-treatment and dried in ultrasound-assisted air dryer.

were tested. A slight reduction in the Biot number was observed for the samples subjected to ultrasonic pre-treatment followed by ultrasound-assisted air-drying, but even in these cases the internal resistance still is the most influential factor in the drying process.

The ultrasonic pre-treatment is known to form microscopic channels inside fruit samples, as has been shown for melons, papaya, pineapples and sapota (Fernandes et al., 2009; Fernandes and Rodrigues, 2008a; Rodrigues et al., 2009). Fig. 2 shows the micrographics of the tissue structure of the dried apple samples. Small microscopic channels have been formed in apples after application of the ultrasonic pre-treatment. The high porosity of apples (compared to other fruits) and the formation of small microscopic channels have influenced in the increase in water effective diffusivity observed when the ultrasonic pre-treatment was followed by conventional air-drying.

Ultrasound-assisted air-drying has provoked the breakdown of cells in the apple tissue, especially at the sample surface. Breakdown of cells is caused mainly by cavitation of small water vapor bubbles that implode due to the expansion and contraction caused by the ultrasonic energy. Ultrasonic application also forms shock waves and jet streaming, which also contributes to breakdown cells and creating microscopic channels in the food structure. These effects leads to a higher increase in the water effective diffusivity in apples. The combined ultrasonic process has increased the breakdown of cells in the apple tissue and consequently the water effective diffusivity has increased even further.

The increase in water effective diffusivity can therefore be correlated with the increase in drying temperature, increase in air velocity and the increase in the breakdown of cells in the apple tissue cause both by ultrasound pre-treatment and by ultrasound-assisted air-drying.

4. Conclusion

The combined ultrasound process comprising the ultrasound pre-treatment and the ultrasound-assisted air-drying increased both internal and external mass transfers of apples cubes, reducing its drying time. The highest reduction in total processing time (132 min) was also obtained applying the ultrasonic pre-treatment followed by ultrasound-assisted convective air-drying when operating at the lowest temperature (45 °C) and lowest air velocity (1 m/s).

The increase in the internal water diffusivity was as high as 93% for the combined ultrasonic process in relation to the conventional air-drying process and the increase in the external mass transfer was as high as 10%. The influence was more intense at lower air velocities and drying temperatures. The highest internal water diffusivity (7.4×10^{-7} m^2/s) was obtained applying the ultrasonic pre-treatment followed by ultrasound-assisted convective air-drying, showing the positive synergy between these two processes.

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Notation

A_S	total surface area of the fruit sample (m^2)
Bi_M	Biot Number (dimensionless)
D	water diffusivity (m^2/s)
K_C	external mass transfer coefficient ($kg/m^2.s$)
V_S	volume of the fruit samples (m^3)
W	moisture content of the fruit ($kg_{water}/kg_{dry\ solids}$)
W_{eq}	equilibrium moisture content of the fruit ($kg_{water}/kg_{dry\ solids}$)
W_{sup}	moisture content at the surface of the fruit ($kg_{water}/kg_{dry\ solids}$)
W_∞	moisture content of the air (kg_{water}/kg)
t	time (s)
δ	bed height of the fruit (m)
ρ	fruit density (kg/m^3)

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