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Mechanisms for improving mass transfer in food with ultrasound technology: Describing the phenomena in two model cases

Alberto Claudio Miano [a], Albert Ibarz [b], Pedro Esteves Duarte Augusto [a,*]

[a] Department of Agri-food Industry, Food and Nutrition (LAN), Luiz de Queiroz College of Agriculture (ESALQ), University of São Paulo (USP), Piracicaba, SP, Brazil
[b] Department of Food Technology (DTA), School of Agricultural and Forestry Engineering (ETSEA), University of Lleida (UdL), Lleida, Catalunya, Spain

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ABSTRACT

The aim of this work was to demonstrate how ultrasound mechanisms (direct and indirect effects) improve the mass transfer phenomena in food processing, and which part of the process they are more effective in. Two model cases were evaluated: the hydration of sorghum grain (with two water activities) and the influx of a pigment into melon cylinders. Different treatments enabled us to evaluate and discriminate both direct (inertial flow and "sponge effect") and indirect effects (micro channel formation), alternating pre-treatments and treatments using an ultrasonic bath (20 kHz of frequency and 28 W/L of volumetric power) and a traditional water-bath. It was demonstrated that both the effects of ultrasound technology are more effective in food with higher water activity, the micro channels only forming in moist food. Moreover, micro channel formation could also be observed using agar gel cylinders, verifying the random formation of these due to cavitation. The direct effects were shown to be important in mass transfer enhancement not only in moist food, but also in dry food, this being improved by the micro channels formed and the porosity of the food. In conclusion, the improvement in mass transfer due to direct and indirect effects was firstly discriminated and described. It was proven that both phenomena are important for mass transfer in moist foods, while only the direct effects are important for dry foods. Based on these results, better processing using ultrasound technology can be obtained.

1. Introduction

Ultrasound technology has been widely studied as an alternative for improving food processing in such operations as defoaming, freezing, extraction, emulsification, hydration, drying and others [1]. In mass transfer unit operations, the ultrasound technology has been successfully used in different processes, such as extraction [2], drying [3–5], osmotic dehydration [6], hydration [7,8], and desalting [9].

The enhancement of the mass transfer unit operation by ultrasound technology has been attributed to different mechanisms. These are considered to be direct and indirect effects of ultrasound. The direct effects are related to the "sponge effect" and inertial flux. When ultrasonic waves travel through the product, they cause a rapid alternating compression and expansion of the tissue matrix, which is compared to a sponge squeezed and released repeatedly [10–12]. This phenomenon can keep micro channels and pores unobstructed, facilitating mass transfer [12]. Further, it can promote mass flow due to pumping. However, although it is frequently attributed with these direct effects on the mass flow [4,9,13–15], these have not been demonstrated during food processing yet.

The indirect effect is related to micro channel formation due to the acoustic cavitation [16]. When ultrasound waves travel through the product, the phenomenon of cavitation takes place in the water inside or outside the product cells, resulting in cell and tissue disruption and the consequent formation of cavities and micro channels. In fact, micro-channel formation due to ultrasound has been shown in various moist foods, such as melons, potatoes, strawberries, apples and cod [3,9,14,17,18]. In addition, the presence of these micro-channels is believed to be the main effect of the ultrasound technology in enhancing the mass transfer phenomena in food processing [6,8,9,19]. However, the formation of micro channels and its importance for mass flow has not been studied for dry foods, such as grain. As the water activity of these products is low, the lower water vapor pressure can limit cavitation, reinforcing the need for evaluation.

Consequently, this work aimed to demonstrate how these mechanisms improve the mass transfer phenomena in food pro-

* Corresponding author at: Avenida Pádua Dias, 11, Piracicaba, SP 13418-900, Brazil.

E-mail addresses: cmiano@usp.br (A.C. Miano), aibarz@tecal.udl.cat (A. Ibarz), pedro.ed.augusto@usp.br (P.E.D. Augusto).

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cessing, and which part of the process they are more effective in. Therefore, two model cases were evaluated: the hydration of sorghum grain and the influx of a pigment into melon cylinders.

2. Materials and methods

The mechanisms of ultrasound enhancement during mass transfer processes were studied in two kind of food: sorghum grain (representing dry foods, with low water activity) and yellow melon cylinders (representing moist foods, with high water activity). Moreover, these two foods have already shown good results when treated with ultrasound during osmotic dehydration [14] and hydration [8].

During the experiments, an ultrasonic bath with a frequency of 40 kHz and a volumetric power of 28 W/L (USC-1400, Unique Brazil) was used. This bath had its piezoelectric elements arranged below the tub. These generated the mechanical waves that are transmitted through the water (or solution), reaching the product. The volumetric power was determined following the method described by Tiwari, Muthukumarappan, O'Donnell and Cullen [20], and it was the same or very close to that used in previous works [8,15]. The temperature of the water was controlled using a stainless steel heat exchanger coupled to an external water bath, which was placed at the top of the solution inside the ultrasonic water bath.

2.1. Ultrasound mass transfer enhancement on grain

For sorghum grain, the hydration process was chosen as the evaluated mass transfer processing.

Sorghum grains (*Sorghum bicolor*) with water activity of 0.653 ± 0.001 and a moisture content of $12.46 \pm 0.17\%$ d.b. (g water/100 g of dry matter) were used. Furthermore, in order to prepare a sample with higher water activity (0.985 ± 0.003) the grains were hydrated for 3 h at 25 °C. Then, these grains were superficially dried and put into sealed containers for two days at 5 °C in order to homogenize the moisture. Consequently, the evaluation was carried out using two different conditions of water activity.

Different treatments were performed in order to identify the mechanism of mass transfer enhancement caused by the ultrasound technology (Fig. 1). These treatments helped to differentiate the indirect effects (micro-channel formation) with the direct effects (the sponge effect, inertial flow), as well as the moment these acted during processing.

Three treatments were performed for the low water activity grains:

- Treatment 1 (TS1: H) consisted of hydrating the grains (15 g of grains in a beaker with 2 L of distilled water) without the application of ultrasound at 25 °C throughout the process (2 h).
- Treatment 2 (TS2: PUS/H) consisted of vacuum packing one layer of sorghum grains in order to treat the grain with ultrasound without it becoming hydrated. This pack was placed at the bottom of the water bath to receive the sound waves better. After 3 h of pretreatment, the grains were unpacked and hydrated (beaker with 2 L of distilled water) without ultrasound application at 25 °C for 2 h.
- Treatment 3 (TS3: H + US) consisted of hydrating the grains (15 g of grains in the ultrasonic bath with 2 L of distilled water) with the application of ultrasound at 25 °C throughout the process (2 h).

Four treatments were performed for the high water activity grains. Treatments 1, 2 and 3 were the same as those applied to the low water activity grains. The other was:

- Treatment 4 (TS4: PUS/H + U) consisted of pretreating the grains, as in treatment 2, but, after that, hydrating them (15 g of grains in the ultrasonic bath with 2 L of distilled water) with the application of ultrasound at 25 °C for 2 h.

During the hydration process, the samples were periodically drained, superficially dried and their moisture content was obtained by mass balance. The sampling was carried out each 15 min for 2 h. All the treatments described above were performed in triplicate. The results were presented as the mean and the standard deviation.

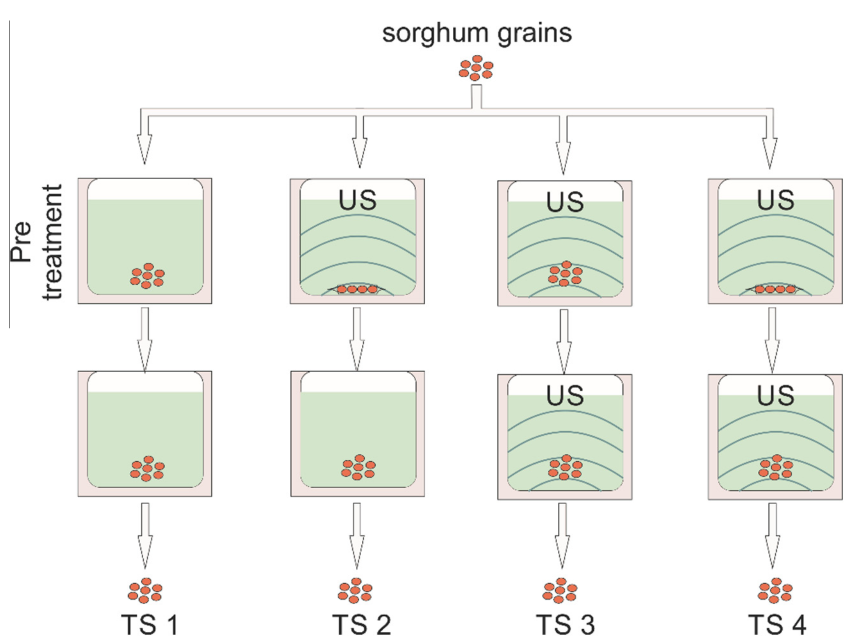

Fig. 1. Treatments to differentiate the ultrasound mechanisms (direct and indirect effects) that enhance mass transfer on the sorghum grains.

2.2. Ultrasound mass transfer enhancement on melon cylinders

The effect of ultrasound processing on the mass transfer phenomena in foods was studied by considering pigment transfer into canary melon (*Cucumis melo inodorus*) cylinders (89.6 ± 0.6%w.b. of moisture, 9.4 ± 0.5 °Brix and 5.74 ± 0.08 of pH). The melon was cut into cylinders in order to obtain homogeneous and uniform specimens.

By evaluating pigment flow into the cylinders in processes under ultrasound, without ultrasound and with a pre-treatment using ultrasound, it was possible to evaluate the direct effects of ultrasound (sponge effect, inertial flow) and those related to the changes in the product microstructure caused by the acoustic cavitation (micro-channel formation; indirect effects).

Melon cylinders of 4 cm long and 1.5 cm in diameter were obtained using a fruit corer. For some treatments (Fig. 2; explanation as follows), the cylinders were perforated using a 0.3 mm-diameter needle. 50 perforations were done randomly along the cylinder, and in the direction of its diameter. The cylinders were perforated in order to simulate higher porosity generated by the micro channels formation and to discriminate the direct effects (i.e., to guarantee samples with micro channels and then evaluate their behavior during processing, in order to compare it with the other treatments).

Ten cylinders were immersed in a beaker or in the ultrasonic bath (depending on the treatment) with 2 L of brilliant blue (food grade, kindly donated by SanLeon, Brazil, www.sanleon.com.br) solution (0.0625 g/L) at 25 °C for the mass transfer evaluation. Six different treatments were considered in order to discriminate the ultrasound effects on the mass transfer process. The treatments are shown in Fig. 2:

- Treatment 1 (TM1: PS) consisted of immersing the cylinders in the pigment solution without ultrasound application throughout the process (2.5 h).
- Treatment 2 (TM2: PS + US) consisted of immersing the cylinders in the pigment solution with ultrasound application throughout the process (2.5 h).
- Treatment 3 (TM3: W/PS) consisted of immersing the cylinders in distilled water without ultrasound application for 60 min as a pretreatment. Then, the process continued in the pigment solution without ultrasound for 1 h.
- Treatment 4 (TM4: W + US/PS) consisted of immersing the cylinders in distilled water with ultrasound application for 60 min as a pretreatment. The process then continued in the pigment solution without ultrasound for 1 h.
- Treatment 5 (TM5: P/PS) consisted of immersing the perforated cylinders in the pigment solution without ultrasound application throughout the process (1 h).
- Treatment 6 (TM6: P/PS + US) consisted of immersing the perforated cylinders in the pigment solution with ultrasound application throughout the process (1 h).

The mass transfer process was evaluated for 2.5 h, removing one cylinder each 30 min for treatments 1–4. For treatments 5 and 6, the mass transfer process was evaluated for 1 h removing one cylinder each 15 min. The removed cylinders were quickly washed with distilled water, drained and superficially blotted with absorbent paper before evaluation. Then, 2 g of the cylinder (after discarding its edges) was triturated with 8 mL of distilled water using a Ultra Turrax homogenizer (IKA® T25D, Brazil) for 30 s at 104 RPM, and then filtered with Whatman grade 2 filter paper. The filtered absorbance at 630 nm of wavelength (maximum absorbance of the pigment solution) was obtained for each cylinder using a spectrophotometer (Femto 600S, Brazil). Following the Beer–Lambert Law, the higher absorbance was considered as the higher pigment solution concentration in the cylinders. All the treatments described above were performed in triplicate. The results were presented as the mean and the standard deviation.

2.3. Micro channels formation observation

In order to evaluate the formation of micro channels due to ultrasound technology, cylinders of agar gel (2%; Oxoid – Thermo Fisher Scientific, Inc. USA) 4 cm long and 1.5 cm in diameter) were considered as model foods. They were treated with ultrasound in 2 L of brilliant blue solution (0.0625 g/L) at 25 °C for 2.5 h (the same time as the melon cylinder treatments). This was carried out to obtain images for better visualization. The cylinders were placed in a black background illuminated from the side with a LED Flashlight and the images were obtained using a regular digital camera.

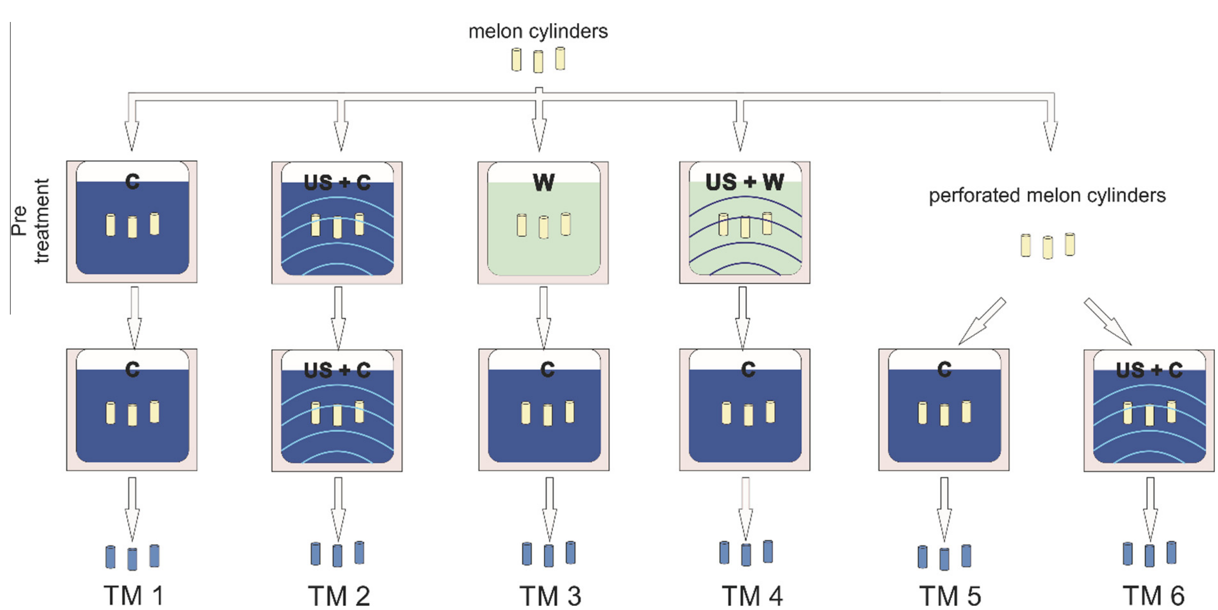

Fig. 2. Treatments for differentiate the ultrasound mechanisms (direct and indirect effects) that enhance mass transfer on the melon cylinders.

3. Results and discussion

3.1. Ultrasound mass transfer enhancement in grains

Fig. 3 shows the application of the ultrasound technology as a treatment and pretreatment to the sorghum grains, representing a food with low water activity. As expected, when the grains were hydrated in the ultrasonic bath throughout the process (TS3), the water intake was improved compared with the conventional hydration process (TS1) [8]. In contrast, when the packaged grains were treated with 3 h of ultrasound and then hydrated without it (TS2), they did not hydrate faster than with the conventional process (TS1). These three treatments demonstrated that ultrasound technology does not generate micro channels in sorghum grains under these conditions of water activity (0.6533 ± 0.0004), or at least, this generation is negligible. The low vapor pressure of the water in the grains hinders cavitation because the cavitation bubbles contain less vapor from the solvent [21]. That is why it is difficult for the micro channels to form inside the product by cavitation. It means that, at this water activity, ultrasound enhanced sorghum grains hydration due to direct effects that caused water to enter by inertial flow and/or the sponge effect.

On the other hand, when the same treatments were performed in sorghum grains with a higher water activity (0.9851 ± 0.0029), different results were obtained. Fig. 4 shows that when ultrasound is applied as a pretreatment (TS2) and the grains are then conventionally hydrated, there is an improvement in the hydration process. Micro-channel formation during the pretreatment was demonstrated, and this improved the mass transfer by reducing internal resistance. In contrast to the low water activity grains, the higher vapor pressure of the water facilitated cavitation and the formation of micro-channels, probably by cell disruption and matrix rupture.

Furthermore, when the hydration of the high water activity sorghum grains was assisted with the ultrasound technology (TS3), there was no improvement in the early stages, but the water intake began to increase with the process time. Although these grains had the right conditions to be affected by cavitation (high water activity) and to form micro channels, a certain process time was necessary for this to be effective. Apparently, acoustic cavitation occurs randomly inside the food matrix, causing the formation of micro cavities, which grow in size with the passing of time. They start to form cavities with different tortuosity and permeability. Many of these cavities can lack connection between each other or with the external medium, thus not improving or only slightly improving the mass transfer. Finally, the mass transfer phenomena are improved when a reasonable number of cavities are formed and/or when there are connections between those and the external medium, forming channels.

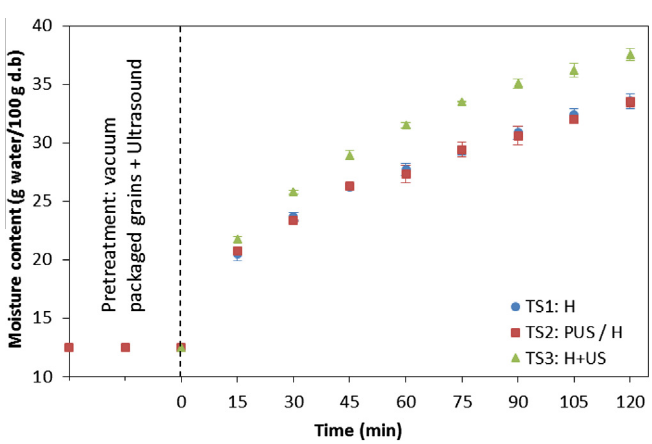

Fig. 3. Evolution of moisture content of the treatments 1, 2 and 3 on the hydration of sorghum grains with low water activity (0.653 ± 0.000) (H: conventional hydration process; PUS: pretreatment with ultrasound; H + US: ultrasound assisted hydration process. The slash means that a pretreatment was used). The dots are the average of the experimental values and the vertical bars are the standard deviations.

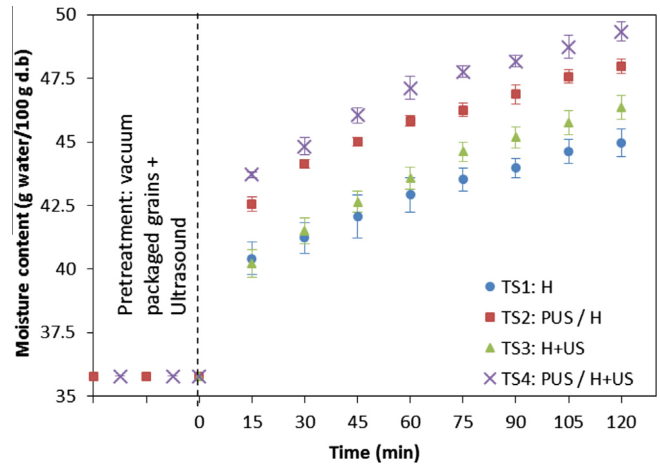

Fig. 4. Evolution of moisture content of the treatments 1, 2, 3 and 4 on the hydration of sorghum grains with high water activity (0.985 ± 0.003) (H: conventional hydration process; PUS: pretreatment with ultrasound; H + US: ultrasound assisted hydration process. The slash means that a pretreatment was used). The dots are the average of the experimental values and the vertical bars are the standard deviations.

Cylinders of agar gel were used to try to see the micro-channel formation, and these were treated with and without ultrasound technology (Fig. 5). The formation of different kind of cavities by ultrasound technology was observed, confirming the statement above (Fig. 6). The different kind of cavities and micro channels formed can have varied tortuosity, permeability and diffusion, improving or not the mass transfer in different ways [22].

Finally, treatment 4 (TS4), which consisted of hydrating the sorghum grains using the ultrasound technology after the pretreatment with ultrasound, showed an even better hydration rate than treatment 2 (TS2), which consisted of hydrating the grains without using the ultrasound technology after the pretreatment with ultrasound. It not only proves that micro channels form, but also the direct effects (inertial flow and "sponge effect") were important for enhancing mass transfer. Moreover, it means that

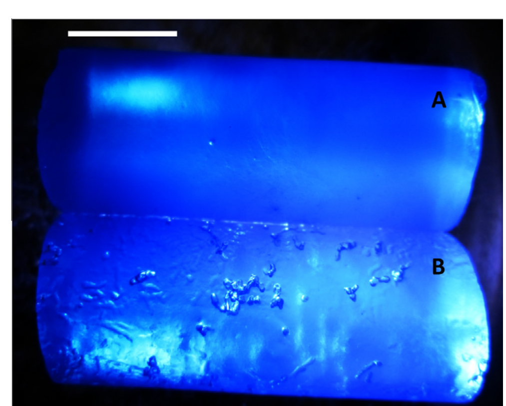

Fig. 5. Agar gel cylinders treated without ultrasound (A) and with ultrasound (B). The white bar is equivalent to 1 cm.

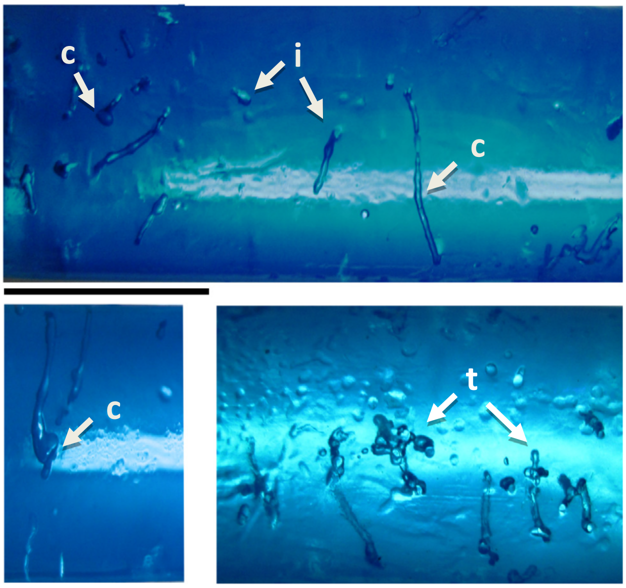

Fig. 6. Different kind of cavities and micro channels: tortuous (t), isolated cavities with lack of connectivity (i), and with external medium connectivity (c). The black bar is equivalent to 1 cm.

the higher porosity of the grain, caused by micro-channel formation, enhance the direct effects of the ultrasound.

Based on these results, ultrasound-assisted hydration of grains can be explained.

When a grain is hydrated, firstly, the enhancement caused by the ultrasound technology is due to the direct effects (inertial flow and "sponge effect"). Then, from a certain water activity, the formation of micro channels starts to take place, further improving mass transfer. This explains why the enhancement of the hydration process is higher after a certain processing time than in the early stages. The few studies of ultrasound assisted grain hydration confirm this [7,8,15], and it can be seen that the differences between the hydration curves grow wider as the process time passes.

Furthermore, these results highlight that the effect of applying the ultrasound technology is probably higher in products with higher water activity. Consequently, the use of this technology in food processing could be designed to maximize its effects, for example, by applying it only at some stages of the process.

Finally, it is important to highlight that the obtained results can only be directly applied for sorghum grains at this level of ultrasonic volumetric power. Each grain has its own structure and composition, which can be differently affected by the ultrasound technology. Further, different volumetric powers can change the relative importance of each mechanism during the ultrasonic processing. Despite this, the main qualitative finds of the present work can be generally interpreted.

3.2. Ultrasound mass transfer enhancement in melon cylinders

Fig. 7 shows the results of the treatments 1 (TM1), which consisted of immersing the melon cylinders in the pigment solution without ultrasound, and 2 (TM2), which consisted of immersing the melon cylinders in the pigment solution with ultrasound. These treatments showed that the ultrasound technology improves pigment intake. It can be clearly noted that the ultrasound almost doubles the pigment retention. Nevertheless, they do not indicate which ultrasound mechanism leads to this improvement.

Fig. 8 shows treatments 3 (TM3), which consisted of pretreating the melon cylinders in water (as a control treatment) before they

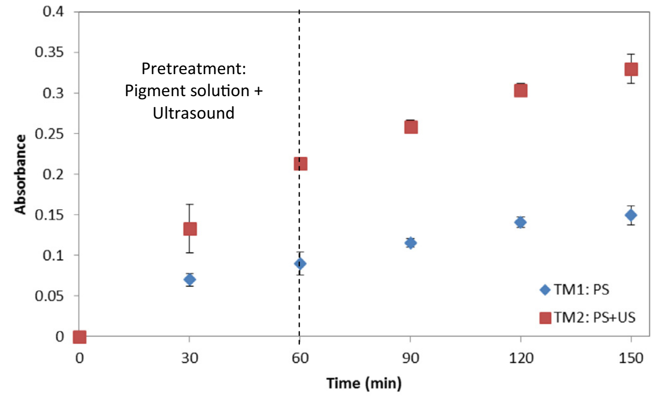

Fig. 7. Evolution of pigment retention of the treatments 1 and 2 the pigment solution transfer on melon cylinders (PS: pigment solution immersion; PS + US pigment solution immersion with ultrasound application). The dots are the average of the experimental values and the vertical bars are the standard deviation.

were immersed in the pigment solution, and 4 (TM4), which consisted of pretreating the melon cylinders in the ultrasonic bath before they were immersed in the pigment solution. They showed that pigment transfer was improved by applying the ultrasound as a pretreatment, demonstrating the formation of micro channels in the melon cylinders. These channels promoted the pigment influx by capillarity, increasing the total concentration of pigment in the melon cylinders. The micro-channel formation was caused by acoustic cavitation, which could cause cell disruption and/or matrix rupture inside the food, generating microscopic channels that reduce the internal resistance to the mass flow [16]. In fact, this mechanism was demonstrated when ultrasound was applied as a pretreatment in melon before its dehydration [19]. Furthermore, it should be mentioned that the formation of micro channels in melon cylinders was probably faster than in the sorghum grains due to the softer matrix and higher water activity of the melon.

Fig. 9 shows treatments 5 (TM5), which consisted of immersing the melon cylinders in the pigment solution without ultrasound, and 6 (TM6), which consisted of immersing the melon cylinders in the pigment solution with ultrasound, both using perforated melon cylinders. Consequently, these cylinders already contained many micro channels before contact with the pigment solution. It can be seen that pigment retention was higher even in the early stages of contact with the solution, where the micro-channel formation by the ultrasound is expected to be low in comparison with those previously generated by the needle. Therefore, this result

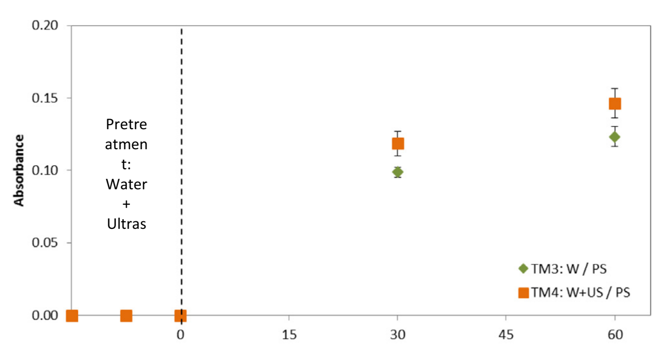

Fig. 8. Evolution of pigment retention of the treatments 3 and 4 for the pigment solution transfer on melon cylinders (W: water immersion; PS: pigment solution immersion; W + US water immersion with ultrasound application. The slash means that a pretreatment was used). The dots are the average of the experimental values and the vertical bars are the standard deviations.

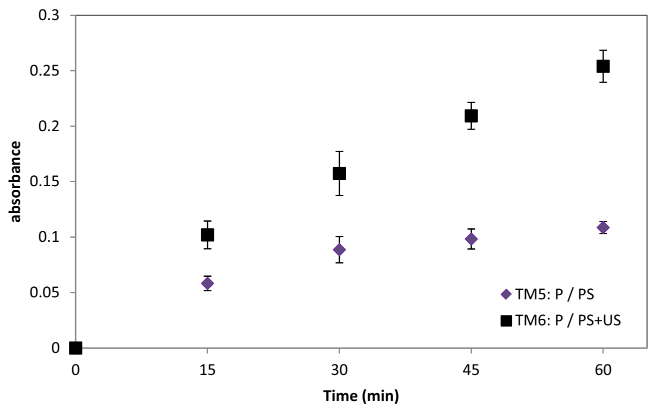

Fig. 9. Evolution of pigment retention of the treatments 5 and 6 for the pigment solution transfer on melon cylinders (P: needle perforation; PS: pigment solution immersion; PS + US pigment solution immersion with ultrasound application. The slash means that a pretreatment was used). The dots are the average of the experimental values and the vertical bars are the standard deviations.

demonstrated that the ultrasound enhancement of the mass transfer also occurs due to direct effects (inertial flow and "sponge effect"). Since the formation of micro-channel by cavitation needs time (due to the random distribution of cavitation until the formation of channels), these treatments were evaluated in only one hour in order to despise subsequent micro-channel formation and thus, evaluate only the direct effects. It can clearly be seen (Fig. 9) that after 15 min, there was an improvement in pigment solution transfer, which means that the direct effect had taken place. Using ultrasound, the pigment flowed into the cylinders faster by the pre formed channels and all the natural cavities. Once they were full with the pigment solution, it was later transferred by diffusion, increasing the total concentration of the pigment inside the cylinders. The ultrasound waves also produced a rapid series of contractions and expansions ("sponge effect") in the melon cylinders, causing the pigment solution to flow through the preformed micro channels, enhancing the mass transfer by reducing internal resistance [11]. It means that the direct effects of ultrasound are enhanced by the porosity of the medium.

Finally, it is important to highlight that the obtained results can only be directly applied for the evaluated pigment influx into melon cylinders at this level of ultrasonic volumetric power. Each component that is transferred (mass transfer) through a food matrix has different behavior since they have different sizes, charge, molecular weight, etc. Therefore, the effect of the ultrasound technology could be different for different components. In addition, since the acoustic cavitation depends on the water vapor pressure, the ultrasound effect (especially the indirect effects) could change due to the water vapor pressure change with the presence of solutes. Further, different volumetric powers can change the relative importance of each mechanism during the ultrasonic processing. Despite this, the main qualitative finds of the present work can be generally interpreted.

3.3. Final considerations

According to the results, the ultrasound-assisted processes could be optimized by taking the water activity and porosity of the food into count. If the aim is to take advantage of the indirect effects of ultrasound technology, for instance, in the dehydration processes, it would be better to use the ultrasound technology as a pretreatment or in the first part of the process since the food has higher water activity. However, in the hydration process, it may be better to use the ultrasound technology after a certain time in the process when the food has a higher water activity. On the other hand, to take advantage of the direct effects of ultrasound technology, it would have to be used on porous food.

It should be mentioned that these results were obtained with a specific ultrasonic volumetric power and frequency (28 W/L; 40 kHz), similar to other works were the ultrasound technology was proven to enhance mass transfer processes in food [8,15,23]. Different result could be obtained with different conditions of power and food products.

The direct effects ("sponge effect" and inertial flow) can be enhanced by the porosity and cavities of the food. The ultrasound waves generate the contraction and relaxation of the tissues causing the fluids be displaced since it acts as a pump. Therefore, spaces in the food are required to the flow of the fluid. The indirect effects (micro channels formation due to cavitation) can be affected by the structure. If the structure is very compact and rigid, the cavitation can take more time to generate cavities, or a higher ultrasound energy would be needed.

However, although the obtained results can only be directly applied for the two model cases here evaluated, the main qualitative finds of the present work can be generally interpreted.

In conclusion, this work can contribute to both academic knowledge and industrial application, since its results can help to optimize the different processes where mass transfer is involved, deciding at which point in the process it is better to use the ultrasound technology and reducing the cost and energy used.

4. Conclusions

This work demonstrated that the ultrasound mechanisms (indirect effect related to micro-channel formation by acoustic cavitation and the direct effects related to the inertial flow and the "sponge effect") enhance the mass transfer in food. Further, those mechanisms can occur in food processing, and their importance is in function of the water activity in the food. Acoustic cavitation takes place more easily in food with higher water activity, resulting in micro-channel formation, while the direct ultrasound effects take place in both low and high water activity food. Moreover, the direct effects are enhanced by the porosity of the food and the micro channels formed. It was also demonstrated that cavitation forms cavities randomly. They are differentiated by their tortuosity and lack of connection with the external medium. Consequently, the micro channels need time to form well, resulting in an improvement in the mass transfer. As a conclusion, based on the results of this work, the application of ultrasound in food processing can be revised in order to maximize its effects, for example only applying it in some periods of the whole process.

Acknowledgments

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