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Additional Information

1 FOOD PROCESS INNOVATION THROUGH NEW TECHNOLOGIES: USE OF ULTRASOUND

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5 **Keywords:** Food processing, mass transport, drying, brining, supercritical extraction

6 **Abstract**

7 The use of new or non-conventional technologies widens the food processing innovation
8 possibilities. Among technologies with a potential application, high intensity ultrasonics has
9 emerged. Ultrasound is a mechanical wave that can affect transport phenomena. Accordingly,
10 the effect associated to ultrasonic application will be dependent on the medium where
11 ultrasound is travelling and on the material to be affected. In this work, ultrasonic applications in
12 different media, such as liquid, gas and supercritical fluid, are addressed as innovative
13 alternatives to enhance transport phenomena and highlight the main factors affecting the
14 process.

15 **1. Introduction**

16 Food processing is in constant evolution in response to different challenges. The changes in
17 consumer tastes and the need to produce safe and high quality foods are responsible for the
18 evolution of the established food processes or the development of the new ones. In this sense,
19 the introduction of new technologies could lead to a reduction of the processing time or an
20 improvement in operating conditions. These aspects are closely linked to the search for high
21 quality products that preserve the natural characteristics of foods. Another important aspect that
22 must be taken into account is the reduction of the energy needs of the processes, thereby
23 decreasing both environmental and financial costs. Ultrasound is an example of new technology
24 and its application in food processing could lead to both these areas undergoing an
25 improvement. On the one hand, ultrasound could be applied as a diagnosis technique to control
26 aspects, food product or processes. On the other hand, ultrasound could be used to improve
27 food processes by affecting the kinetics, the yield or the product quality. This work will focus on
28 the latter.

29 Acoustic waves are mechanical waves that need a material medium to propagate. Usually, they
30 are classified by taking the human audible frequency as reference. This range could be placed
31 between 20 Hz to 20 kHz. Lower frequencies are referred to as infrasound and higher
32 frequencies as ultrasound. The ultrasound waves suffer changes in their properties (velocity,
33 attenuation, frequency spectrum,...) when travelling through a medium. The study of these
34 variations is used in diagnosis applications to characterize the medium. In these applications,
35 the frequency of the waves is in the range of MHz and the power applied is not higher than 1
36 W/cm² (Patist and Bates, 2008). When the applied power of ultrasound is higher, the acoustic
37 waves could affect the medium generating interesting effects for industrial applications. This use
38 of ultrasonic technology is known as "power ultrasound" or "high intensity ultrasound" and the

39 main objective is to induce changes in products or processes. In this case, the frequency is in
40 the range of 20-100 kHz (Mason and Lorimer, 2002).

41 The aim of this work was to address different power ultrasound applications in order to highlight
42 factors affecting some innovative approaches to food processing.

43 **1.1. Effects of ultrasound**

44 The effects produced by high power ultrasound when travelling across a medium are diverse
45 and their relative importance depends on the characteristics of the medium. In general,
46 ultrasound produces alternating compression and decompression of the media. In liquids, when
47 ultrasonic power attains a threshold, the rarefaction cycle may exceed the attractive forces and,
48 from existing gas nuclei, cavitation bubbles could appear (Soria and Villamiel, 2010). These
49 bubbles could maintain a stable increasing and decreasing size giving rise to the so-called
50 "stable cavitation" generating a micro-agitation of the medium. However, the bubbles can also
51 grow and collapse generating very high local temperatures (5000 K) and pressures (1000 atm),
52 which produce, in turn, high energy shear waves and turbulence in the cavitation zone. This last
53 effect is known as "transient cavitation" (Leighton, 1998). The implosions are asymmetric if
54 produced near a solid surface generating a microjet that hits the solid (Mason, 1998). This is the
55 main effect observed in the use of high intensity ultrasound in cleaning operations. Moreover,
56 the microjets hitting the solid food surface may produce an injection of fluid inside the solid
57 (Mason and Cordemans, 1996). The intensity of cavitation and its effects depend on the
58 characteristics of the medium, such as viscosity, and/or process variables, like ultrasonic
59 intensity, ultrasonic frequency or pressure.

60 In gas media, the main challenge of the application of ultrasound is attaining an efficient
61 transmission of the acoustic waves at high frequencies due to the acoustic impedance
62 mismatch between transducers and gas as well as the high ultrasonic attenuation in gas media.
63 However, when ultrasound is applied in an efficient way, it can produce intense effects on the
64 interfaces, such as pressure variations or microstirring, which can affect the mass transfer
65 phenomena (Carcel et al., 2007a) by reducing the boundary layer thickness.

66 In solid materials, alternative compressions and expansions generated by the ultrasonic waves
67 produce a similar effect to that observed when a sponge is squeezed and released repeatedly
68 (De la Fuente et al., 2006). This "sponge effect" produces the release of liquid from the inner
69 part of the particle to the solid surface and the entry of fluid from outside. The forces involved in
70 this mechanism can be higher than the surface tension which maintains the water molecules
71 inside the capillaries of the material, creating microscopic channels (Muralidhara et al., 1985)
72 and making the interchanges of matter easier. Other effects to be considered are the variation
73 of viscosity, surface tension or the deformation/degradation of the solid structure.

74 From a general point of view, all the effects produced by ultrasound could influence mass
75 and/or heat transfer phenomena. In treatments with a solid immersed in a fluid, ultrasound could
76 accelerate the internal transport making the entry of fluids in the solid matrix and/or their exit

77 easier and also facilitating the exchanges between the solid surface and the surrounding fluid.
78 Then, the use of ultrasound, when applied in an efficient way, could be interesting in
79 applications involving heat or mass transport, decreasing both the external and internal
80 resistance to transport.

81 **1.2. Applications of high intensity ultrasound in food processing**

82 The main applications of ultrasound in food processes are linked to the effects it has on heat or
83 mass transfer operations. Most of the ultrasonic applications reported in literature are found in
84 liquid-liquid and liquid-solid systems (Mulet et al., 2003) due to the relative ease with which
85 ultrasonic waves are transmitted in liquids. There is a wide offer of commercial equipment
86 available on the market, including ultrasonic baths and different probe systems, which may be
87 adapted for different operations. Thus, ultrasound has been applied in osmotic dehydration
88 (Cárcel et al., 2007b; Fernandes and Rodrigues, 2007; Jambrak et al., 2007), brining, (Cárcel et
89 al., 2007c; Gabaldon-Leiva et al., 2007; Siró et al., 2009), freezing (Delgado et al., 2009),
90 extraction (Vilkhu et al., 2008; Soria and Villamiel, 2010) or enhancement of heat transfer in
91 heat exchangers (Gondrexon et al., 2010). Emulsions are also a field of interest, for example in
92 the production of mayonnaise (Mason, 1998) or traditional products like Xixona turrón (Mulet et
93 al. 1999).

94 The applications in gas-solid systems are less common, because, as mentioned before, the
95 high impedance mismatch and the high ultrasonic energy attenuation in air makes the
96 transmission of ultrasound from the transducer to the air and from the air to the solid difficult
97 (Garcia-Perez et al., 2009). Nevertheless, some applications have been developed in the
98 convective drying field to overcome these challenges. This is the case of the stepped plate
99 ultrasonic transducers developed by Gallego-Juárez et al. (1999). These prototypes have been
100 used in the convective drying of several food products, applying airborne ultrasound or with
101 direct contact between the transducer and the solid (De la Fuente et al., 2006; Gallego-Juárez
102 et al., 2007). Another alternative consists of the development of vibrating drying chambers to
103 apply air borne ultrasonic energy (García-Pérez et al., 2006a). The promising results of this
104 system will be addressed in the following sections.

105 Ultrasonic applications in supercritical media are also scarce (Riera et al., 2004). The use of a
106 supercritical fluid as solvent in the extraction operation has been receiving increasing attention
107 due to its advantages when compared to the conventional extraction processes, such as the
108 product quality or the use of a non-toxic, recyclable, cheap, relatively inert and non-flammable
109 solvent. The main disadvantage of the process is the slow kinetics. Due to the high pressure
110 needed to achieve and maintain the supercritical phase of the solvent, it appears to be difficult
111 to introduce some agitation system inside the extractor. The effects produced by ultrasound
112 (compression and decompressions, radiation pressure, high turbulence, etc.) could increase the
113 extraction kinetics. In the literature, some works have approached this problem by locating the
114 ultrasound application system outside the extractor (Balachandran et al., 2006) and others have
115 succeeded in introducing the transducer inside the extractor (Riera et al., 2002)

116 However, the potential applications mentioned should be examined case by case, because it is
117 not only the medium, solid, liquid, gas or supercritical, but also the process variables
118 (temperature, flow regime, intensity, etc.) and the product structure which could affect the
119 magnitude of the changes induced by ultrasound. These aspects, deriving from applications of
120 ultrasound in different media (liquid-solid, gas-solid and supercritical-solid), will be illustrated.

121 **2. Solid-liquid systems**

122 **2.1. Equipment**

123 The application of ultrasound requires a system capable of producing a stable and reliable
124 ultrasonic field from another type of energy, usually electrical. The transducers are the devices
125 used to convert the energy, coming from a power generator, into mechanical energy in the form
126 of ultrasonic vibrations. There are two main types of transducer: magnetostrictive and
127 piezoelectric. The first, constructed from high-strength metallic alloys, has the advantage of
128 being able to reach high levels of acoustic power intensity, over 150 W/cm^2 , is very stable,
129 reliable and does not age (Peshkovsky and Peshovsky, 2010). However, the relatively low
130 efficiency (below 50%) when compared to piezoelectric systems (up to 95 %), the other type of
131 transducer, is probably the main reason why the latter is more widely used, regardless of the
132 relatively low levels of acoustic power intensity and the short life-span.

133 The transducers are attached to the vibrating system whose function consists of transmitting the
134 vibration from the transducer to the medium. In liquid applications, the most commonly used
135 systems are baths and probe-type systems. In the ultrasonic baths, several transducers,
136 vibrating in phase, are attached to the bottom of a metallic tank transmitting the vibration to the
137 contained liquid. Due to the reflection of ultrasonic waves in the air-liquid interface, a stationary
138 field, with maximum and minimum acoustic intensity zones, is created inside it (**Figure 1**). Then,
139 the applied ultrasonic treatment can change depending on the location of the samples.

140 In the probe systems, ultrasound is directly applied by a vibrating "horn". Depending on the
141 geometry of the probe, it could be used simply to transmit the ultrasonic energy or to
142 concentrate it on a lower surface in order to amplify the intensity and, therefore, their effects
143 (Mason, 1998). In applications with this type of systems, the distance between the sound tip and
144 the treated sample is an important parameter to be controlled due to the attenuation of the
145 ultrasonic field with the distance.

146 **2.2. Influence of ultrasound on the transport resistance**

147 The effects produced by ultrasound in solid-liquid systems could affect the transport process
148 reducing the external transport resistance. Carcel et al. (2004) addressed the influence of
149 ultrasound on convective heat transport by introducing an aluminum cylinder in one ultrasonic
150 cleaning bath (Fungsonics mod. 28 L, 20 kHz, Fungilab S.A., Barcelona, Spain) containing hot
151 distilled water (28 L). The dimensions of the cylinder were chosen in order to neglect the
152 internal resistance to heat transfer. The evolution of the temperature inside the cylinder was
153 logged until there was less than $1 \text{ }^\circ\text{C}$ difference when compared with the bath temperature. Four

154 types of heating tests were carried out: without any agitation of the bath water, with (USWAG)
155 and without ultrasound application (WAG), and agitating the bath water, with (USAG) and
156 without ultrasound application (AG). The experimental results were modelled considering
157 Newton's law of heating and the convective heat transfer coefficient (h) was identified. This
158 model was adequate for describing the heating process, as confirmed by the close agreement
159 between the experimental and calculated temperatures (**Figure 2A**). The h coefficient was
160 significantly different ($p < 0.05$) for the four kinds of experiments carried out and the identified
161 values varied according to: $WAG < USWAG < USAG < AG$ (**Figure 2B**). Therefore, the ultrasound
162 treatment (USWAG) resulted in an h coefficient lower than that of a well-stirred medium (AG)
163 but higher than that of the static condition experiments (WAG). That means that the application
164 of ultrasound reduced the external resistance to heat transfer compared to natural convection.
165 The limited effects on the h values compared to the mechanical stirring of the medium, could be
166 explained by the low acoustic power provided by the ultrasonic bath systems, in order to avoid
167 cavitation damage to the tank walls, and the low power density applied, because there is
168 generally a large volume of liquid in the tanks (Mason, 1998).

169 It must be highlighted that the h coefficient identified in experiments with the simultaneous
170 application of agitation and ultrasound (USAG) presented an intermediate figure between
171 USWAG and AG treatments. This fact could indicate the existence of an interaction between
172 ultrasonic and mechanical agitation resulting in a reduction of the turbulences in the medium. As
173 **Figure 1** shows, the agitation of the medium affected the ultrasonic field decreasing both the
174 average acoustic pressure and the difference between maximum and minimum pressure zones.
175 In this sense, the h value of USAG experiments will indicate that the ultrasonic field could also
176 affect the agitation of the medium, decreasing the turbulence level created.

177 The level of acoustic intensity applied and the type of material treated could influence the
178 magnitude of the ultrasound effects in the transport process. In this sense, it is important to take
179 into account that the treated medium could affect the transmitted acoustic field. As can be
180 observed in **Table 1**, the same ultrasonic system, working under the same conditions, produces
181 different acoustic pressure when in brine than when in sucrose solution and, therefore, the
182 ultrasonic effects can be different. On the other hand, the effects of ultrasound on the mass
183 transport kinetics might not appear until an acoustic intensity threshold is attained. Studying the
184 mass transport of moisture and solutes during the osmotic treatment of apple in a sucrose
185 solution (30 °Brix; 30 °C), Carcel et al. (2007b) found an intensity threshold of 10 W/cm^2 , below
186 which no influence of ultrasound was observed. When the applied ultrasonic intensity was 11.5
187 W/cm^2 , the identified effective diffusivity increased by 117% for moisture transport and 137% for
188 the dry matter transport, compared to the treatments without ultrasound application. This
189 ultrasonic intensity threshold could vary for different products or transport processes. Carcel et
190 al. (2007c) reported that intensity thresholds of 39 and 51 W/cm^2 were needed in order to
191 observe some effects on the moisture or salt transport respectively during pork meat brining
192 (saturated NaCl brine, 2 °C). Above this threshold, the higher the level of applied ultrasonic
193 intensity, the more the ultrasound was observed to affect mass transport.

194 However, the level of acoustic intensity can also affect the type of influence on the mass
195 transport. The levels of acoustic intensity used by Carcel et al. (2007b) (11.5 W/cm^2) in the
196 osmotic dehydration of apples increased the two main mass transport processes that took place
197 in these treatments: moisture loss and solute gain, but the higher levels used by Carcel et al.
198 (2007c) in meat brining ($> 51 \text{ W/cm}^2$) not only affected the kinetics of transport but, in the case
199 of moisture, the transport direction. The samples treated ultrasonically at the higher acoustic
200 intensities tested (75.8 W/cm^2) did not undergo a dehydration process like the conventionally
201 brined meat, but had a higher moisture content than fresh meat. Moreover, the NaCl content of
202 the ultrasonically brined samples after 45 min of treatment was 115% higher than conventionally
203 brined samples. That means that the conventional brining process produced a water loss and a
204 NaCl gain while, at these intensity levels, the application of ultrasound, induced the gain of both
205 water and NaCl. The influence of ultrasound on apple treatments can be explained by ultrasonic
206 effects such as the "sponge effect" or the creation of microchannels, which can affect the
207 internal mass transport resistance, and the generation of microstirring or cavitation, which affect
208 the external resistance by reducing the boundary layer of diffusion. In the case of meat brining,
209 the highest applied acoustic intensity generates a more intense cavitation in brine. The
210 asymmetric implosion of cavitation bubbles near the meat surface produces the formation of
211 microjets that hit the solid (Mason & Lorimer, 2002) and could produce the microinjection of
212 brine into the meat samples. This fact could not only explain the increase in NaCl content but
213 also the increase in sample water content.

214 **3. Solid-gas systems**

215 **3.1. Equipment**

216 As already mentioned, the main drawback of the application of ultrasound in gas media is the
217 transmission of the acoustic wave from the emitter's surface to the samples. The air is a high
218 attenuating medium that absorbs the acoustic energy preventing its transfer to the solids to be
219 treated. On the other hand, the high impedance difference between the solid surface of emitters
220 and the air, and between the air and the solid samples, produces the reflection of a high
221 proportion of the generated acoustic signal (García-Pérez et al. 2006a). This is the reason why
222 there are very few research groups working on the application of ultrasound in food drying. The
223 applications of ultrasound during osmotic pre-treatments prior to the air drying process may be
224 mentioned (Fernandes & Rodrigues, 2007), but these processes are applications in solid-liquid
225 systems. Therefore, it is of great importance to gain further a more thorough knowledge of the
226 mechanisms of the ultrasonic wave transmission in gas media in order to optimize the
227 application systems (de la Fuente et al., 2006). Significant attempts have been made to alleviate
228 these problems by developing a powerful source of airborne ultrasound that can achieve a more
229 efficient transmission of energy to the material.

230 The systems of sirens and whistles convert the kinetic energy of a fluid into an acoustic wave. In
231 sirens, the fluid is forced to pass across a hole, thus generating turbulence that constitutes a
232 mechanical wave. In whistles, the fluid is forced across a thin blade which causes the blade to

233 vibrate. For each vibrational movement, the leading face of the blade produces a pressure wave
234 (Mason, 1998). In liquid application, the whistle constitutes a powerful tool for mixing and
235 homogenization (Mason & Lorimer, 2002). Da Mota and Palau (1999) used a siren system to
236 improve onion drying. A low frequency (1.6 and 3.2 kHz) was used in these experiments to
237 partially avoid the acoustic energy attenuation; this action, however, may involve an intense
238 noise that could be an obstacle to its use.

239 Another group of ultrasonic systems includes the use of a piezoelectric transducer attached to
240 different types of emitters trying to adapt the signal in order to achieve a good transmission to
241 the air. One of the main types is the stepped plate emitters, characterized by a surface emission
242 with a stepped profile that is responsible for the best impedance match with air, the increase in
243 the power capacity of the system and which avoids the phase cancellations produced in flat
244 plate radiators (Gallego-Juárez et al., 1999; Gallego-Juárez, 2010). Circular and rectangular
245 prototypes have been developed for the 10-40 kHz frequency range and power capacities of
246 about 100 W and have been applied in forced air dehydration assisted by airborne ultrasound
247 and also in the direct coupling of the ultrasonic vibrator and the solid matrix. Drying experiments
248 have been carried out on different vegetables such as carrots, potatoes, and mushrooms (De la
249 Fuente et al., 2006; Gallego-Juárez et al., 2007). The drying process which involved direct
250 contact between the vibrating elements and the materials being dried showed a very intense
251 effect which can increase when a low static pressure is applied. The effect that power ultrasound
252 had on drying was reduced when the application was carried out using an airborne technique.
253 The better transmission of vibrations to the sample in direct contact experiments is what is
254 mainly responsible for this fact. Nevertheless, it is very difficult to adapt the direct contact
255 systems to work on an industrial scale. An alternative consists of considering the drying
256 chamber itself as the vibrating element to transmit the acoustic waves to the samples. To apply
257 this concept, García-Pérez et al. (2006a) replaced the drying chamber of a conventional
258 laboratory hot air drier (Sanjuan et al., 2003) by an aluminium vibrating cylinder (internal
259 diameter 100 mm, height 310 mm, thickness 10 mm) driven by a piezoelectric composite
260 transducer capable of generating a high-intensity ultrasonic field inside the cylinder. The driving
261 transducer consists of an extensional piezoelectric sandwich element together with a
262 mechanical amplifier (**Figure 3**). The whole has to be resonant at the frequency of the selected
263 vibration mode of the chamber, 21.8 kHz. The average sound pressure level inside the chamber
264 in stagnant air conditions was 154.3 dB, measured for an electrical power applied to the
265 transducer of 75 W. Therefore, a high intensity acoustic field inside the chamber is obtained with
266 relatively low applied electric power. This system has been used in the drying of different
267 products, such as carrot (García-Pérez et al. 2008), persimmon (Cárcel et al, 2007a), lemon
268 peel (García-Pérez et al., 2009) or olive leaves (Cárcel et al. 2010) and, despite the fact that the
269 vibration transmission transducer-sample is not as good as the direct contact application, the
270 results of the increase in the drying rate have been promising.

271

272 3.2. Influence of process variables.

273 The use of sonication to improve the dehydration process dates from the middle of the 20th
274 century, promoted by the interest in the drying of heat-sensitive materials (Boucher, 1953) due
275 to the limited heating effect of ultrasound on gas systems. Borisov and Ginkina (1973) reported
276 a series of experiments carried out in the Academy of Science of the USSR to determine the
277 influence of the main process variables using fluid driven transducers. Due to the development
278 of more efficient devices with which to apply ultrasound in gas systems, especially the
279 aforementioned vibrating drying chamber, studies into several food materials considering
280 different process variables have been carried out in order to address their influence on the
281 drying rate.

282 In general, the application of ultrasound during drying increases the kinetics of dehydration,
283 affecting both the internal and the external resistance. As can be observed in **Figure 4**, the
284 application of ultrasound in the drying of carrot and lemon peel increased the effective diffusivity
285 and the mass transfer coefficient. The influence on the effective diffusivity could be attributed to
286 the "sponge effect" or the creation of internal microchannels that make it easier for the water to
287 be released from the solid samples. In cryo-SEM observations, Ortuño et al. (2010) found that
288 ultrasonically dried orange peel albedo showed a more compressed cellular structure with larger
289 intercellular air spaces than conventionally dried samples. The alternating expansions and
290 compressions produced by ultrasound created a highly porous material that facilitated the water
291 movement. These authors also found that ultrasound affected the flavedo structure. The
292 conventional air drying process scattered the waxy components, closing the pores and creating
293 a waterproof barrier. Nevertheless, the original ring-shaped waxy accumulations in the pores
294 continued to be well defined. On the contrary, in the samples dried using ultrasound application,
295 these ring-shaped accumulations disappeared revealing the very intense effect that ultrasound
296 had in the interface. The influence of ultrasound on the external resistance to mass transfer
297 could be linked to the generation of differential pressures and the microstirring at the interfaces
298 and these effects should also affect the surface of the treated solid.

299 The magnitude of the effects of ultrasound depends on the process variables, such as air
300 temperature, air velocity, mass load density, applied acoustic energy or the raw material
301 processed. The air velocity has been found to be one of the most important variables involved in
302 power ultrasound assisted air drying (Cárcel et al., 2007a; García-Pérez et al., 2007). From
303 experimental measurements, Riera et al. (2011) found that the increase of the air velocity
304 produced the reduction of the sound pressure level in the drying chamber. As a consequence,
305 the energy available for the samples at high air velocities could not be enough to affect the
306 mass transfer process.

307 The magnitude of the effects of ultrasound on the drying rate also depends on the ultrasonic
308 power level applied. As can be seen in **Figure 4**, an ultrasonic intensity threshold can be
309 achieved to find some evidence of the effect of ultrasound on the kinetic parameters of the
310 process. Above this threshold, the more power is applied, the higher the values of diffusivity and

311 mass transfer coefficient that can be obtained and, therefore, the faster the drying process
312 (García-Pérez et al. 2009). Above the threshold, the relationship between the kinetic parameters
313 and the applied ultrasonic power was linear for the whole range tested.

314 The influence of the power ultrasound also depends on the material to be processed. The
315 structure of the material could be an important factor in the extension of the effects of
316 ultrasound. This fact is also illustrated in **Figure 4**. The influence of ultrasound on diffusivity and
317 mass transfer coefficient appeared in the case of carrots when over 20-30 W power was applied,
318 whereas when drying lemon peel, it can even be observed at the lowest power level tested.
319 Moreover, the slope of the linear relationship between kinetic parameters and the applied power
320 was nearly one order of magnitude higher for lemon peel than for carrot. Lemon peel is more
321 porous than carrot (García-Pérez et al., 2007) and so the expansions and contractions (sponge
322 effect) produced by ultrasound may be more intense due to its low mechanical resistance; in
323 addition, the effects on the interfaces would be more intense because of the large porous
324 volume. Furthermore, a greater absorption of acoustic energy would be expected in high
325 porosity products, thus increasing the energy available in the particle to affect mass transfer
326 processes.

327 The relative effect of ultrasound depends on the transfer resistance affected. If the resistance is
328 low, the effect of ultrasound application is also low. For that reason, the effect of ultrasound
329 application is more evident when drying is carried out at moderate temperatures. For example,
330 when carrying out experiments of the drying of carrot samples at different air temperatures
331 García-Pérez et al. (2006b) found that the influence of ultrasound on the diffusivity was different
332 depending on the temperature. The application of power ultrasound significantly increased
333 ($p < 0.05$) the effective moisture diffusivity at temperatures lower than 60 °C but was almost
334 negligible at 70 °C. A similar influence of the temperature on the effects of ultrasound was found
335 by Gallego-Juárez et al. (1999).

336 The effect of mass load density can be observed in **Figure 5** for drying experiments performed
337 on carrot cubes. In conventional air drying processes, it appears that, the increase of mass load
338 density, in the range tested, does not affect the effective moisture diffusivity although it does
339 produce a reduction of the mass transfer coefficient. This could be linked to perturbations in the
340 air flow through the drying chamber, thus creating preferential pathways and, as a
341 consequence, increasing external mass transfer resistance (Cárcel et al., 2011). When high
342 intensity ultrasound was applied, the mass transfer coefficient and the effective moisture
343 diffusivity increased when the mass load density used was moderate, although the acoustic
344 influence on mass transfer coefficient became negligible at high mass load densities.

345 Therefore, from previous results, for a specific ultrasonic application it appears to be important
346 to carry out a study into the influence of different process variables in order to find the optimum
347 drying conditions. The innovation of the process could fail if not properly addressed.

348

349 4. Solid-supercritical systems

350 Ultrasound has been used extensively in the last two decades as an efficient extraction tool for
351 food engineering purposes and nowadays it has even become the reference for other extractive
352 technologies under development. Ultrasound assisted extraction has been applied in the
353 extraction of valuable compounds, like different herbal extracts, polyphenols, anthocyanins,
354 aroma compounds, polysaccharides or proteins (Vilkhu et al., 2008). The sonication in solid-
355 liquid systems could improve the extraction efficiency and rate, reduce the temperature needed
356 and save solvents thus favouring the solubilisation of the interesting compounds. However,
357 ultrasound assisted extraction is not only related to pure diffusion processes, in fact, the
358 cavitation of bubbles generated by the application of ultrasound in an elastic medium can
359 implode at the surface of the plant and destroy the plant cells (Veillet et al, 2010). However, it
360 frequently presents similar drawbacks to the conventional extraction processes: the use of toxic
361 solvents dangerous both for the environment and for the final quality of the product.
362 Supercritical fluid extraction has become a promising technique with which to solve these
363 problems due to the fact that the solvent commonly used, CO₂, is non-toxic, recyclable, cheap,
364 relatively inert and the process improves the product quality and product recovery (Lang & Wai,
365 2001). However, supercritical fluid extraction presents slow extraction kinetics even when solute
366 free solvent is recirculated and, therefore, improvements in mass transfer are required (Berna et
367 al., 2000). The use of high-intensity ultrasound represents a potentially efficient way of
368 enhancing mass transfer processes (Riera et al., 2004) and, consequently, innovating the
369 supercritical fluid extraction techniques.

370 4.1. Equipment

371 Supercritical CO₂ fluid extraction takes place inside a reactor under high pressure conditions
372 (pressure above 72 bar). This fact makes it difficult to introduce a mechanical agitation system
373 in the reactor, as well as an ultrasonic transducer, which represents the main obstacle to the
374 application of ultrasound in this kind of process. To address it, two different set-ups may be
375 mentioned. One of them consists of the use of a commercial sound probe joined to the wall of
376 the extractor. As the transducer is fitted externally, it is expected that there will be a power
377 attenuation as the ultrasound passes through the stainless steel vessel walls (Balachandran et
378 al., 2006).

379 The other set-up consists of the introduction of the transducer inside the reactor. This solution,
380 proposed by Riera et al. (2002), is based on a piezoelectric sandwich transducer designed and
381 built for this application and inserted in the upper part of the vessel with a 100 W power
382 capacity. The transducer is driven by an electronic generator, which incorporates a system to
383 follow the resonance frequency (**Figure 6**). This is an essential device due to the changes that
384 different process conditions may provoke in the characteristic impedance of the supercritical
385 fluid (Riera et al. 2004). In fact, these authors achieved stable operation conditions when the
386 values of density, pressure and temperature were kept at the operational values. Moreover, any
387 change in the flow rate or density was immediately detected and followed by the control system

388 of the transducer. Therefore, it is possible to use this control system to monitor the extraction
389 process.

390 **4.2. Influence of ultrasound on supercritical fluid extraction**

391 The application of ultrasound during supercritical fluid extraction affects both the kinetics and
392 the yield. Balachandran et al. (2006) studied the application of ultrasound in the supercritical
393 extraction of pungent compounds from ginger using a commercial probe system externally
394 attached to the extractor. The nominal power used was 300 W but, due to the ultrasonic
395 transducer being outside the extractor, the reflection and adsorption of the acoustic wave
396 decreased the actual intensity received by the samples. However, they found an important
397 increase in yield when ultrasound was applied. The particle size could be an important factor to
398 take into account, in fact, the effects of ultrasound increased when the particles were smaller.
399 Thus, for a particle size of 4 mm, the yield was 30 % higher in experiments carried out with
400 ultrasound application compared to non-ultrasonic experiments. Similar behavior was found by
401 Riera et al. (2004) working on almond oil extraction. They used a system where the ultrasonic
402 transducer was inside the reactor and found that ultrasound had a greater influence on the yield
403 at the lowest particle size tested (3-4 mm compared to 9-10 mm), achieving an increase of 20 %
404 in oil recovery. It is likely that the highest surface area to volume ratio favors the action of
405 ultrasound, pointing to an influence of ultrasound on the external resistance. From microscopic
406 pictures (field emission scanning electron microscopy, FESEM), Balachandran et al. (2006)
407 observed that the structure of the material treated with ultrasound showed cellular damage that
408 could favor the removal of the cell contents. This fact could indicate that the effect of ultrasound
409 could also be located in the internal resistance to mass transfer.

410 The influence of ultrasound may also be important in the extraction kinetics. Riera et al. (2004)
411 report a 30 % reduction of the extraction time for a similar extraction yield and found that the
412 influence of ultrasound was more evident in the second period of extraction, after the period
413 when the solubility of the solute in the solvent controls the extraction. On the contrary,
414 Balanchandran et al. (2006) found that ultrasound had the greatest influence on the extraction
415 kinetics in the first phase of extraction, doubling the effective diffusivity identified for a
416 conventional process. The ultrasound enhancement in the second stage of extraction, although
417 significant, was lower than in the first extraction phase. These results show that, the magnitude
418 of the influence of ultrasound on extraction could be different for each specific application.

419 The effects of ultrasound could be related to the compressions and decompressions, the
420 radiation pressure or the streaming (Riera et al., 2004). No clear evidence of cavitation was
421 found under the conditions used for supercritical fluids (Balachandran et al., 2006). The high
422 pressure needed to achieve supercritical conditions, above 72 bar, makes the appearance of
423 cavitation bubbles difficult.

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425

426 **5. Conclusions**

427 The application of ultrasound affects the heat and mass transport processes. The effects linked
428 to ultrasound include cavitation, compressions and expansions, microstirring, etc. and affect
429 both the external and the internal heat and mass transfer resistance. The importance of each
430 effect in the global influence of ultrasound on transport is different for the system considered:
431 solid-liquid, solid-gas or solid-supercritical, since, for example, cavitation does not take place in
432 a gas or supercritical medium. The process variables influence the magnitude of the ultrasound
433 effects and it is necessary to establish the optimum value for each specific application. This
434 offers new possibilities for food process innovation, ranging from energy savings to process
435 yield or product quality. The use of ultrasound is opening up a field of activity in food
436 processing.

437 **6. Acknowledgements**

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- 557

558

559 **TABLE CAPTION**

560

561 **Table 1.** Acoustic pressure measurements (bar) carried out using a hydrophone in 800 mL of a
562 saturated brine and a sucrose solution (30 °Brix). Ultrasound applied with a probe
563 system (probe diameter of 13 mm and at 1.5 cm distance from the emitter's surface)
564 supplying different percentages of the total electric power of the equipment (100 W).

565

566

567 **FIGURE CAPTIONS**

568 **Figure 1.** Variation of acoustic pressure inside an ultrasonic bath (Fungsonics mod. 28 L, 20
569 kHz, Fungilab S.A., Barcelona, Spain) containing distilled water (28 L) with the distance from
570 the bottom. Measurements carried out with and without water agitation.

571 **Figure 2. A.** Evolution of experimental and calculated temperature on an aluminum cylinder
572 during the heating in a bath with ultrasound application. **B.** Identified heat transfer coefficient,
573 h , for heating treatments with agitation (USAG with ultrasound application and AG without)
574 and without agitation (USWAG with ultrasound application and WAG without) of heating
575 medium.

576 **Figure 3.** Detail of the ultrasonic application system of an ultrasonically assisted convective
577 drier (Cárcel et al., 2007a).

578 **Figure 4.** Identified effective diffusivity (D_e) and mass transfer coefficient (k) for the drying of
579 carrot and lemon peel with ultrasound application at different acoustic powers. Air
580 temperature of 40 °C and air velocity 1 m/s (García-Pérez et al. 2009).

581 **Figure 5.** Mass transfer coefficient (k) and effective moisture diffusivity (D_e), identified during the
582 drying of carrot cubes at 40 °C and 1 m/s with (US; 75 W, 21.7 kHz) and without (AIR)
583 ultrasound application (Cárcel et al., 2011).

584 **Figure 6.** Scheme of the supercritical fluid extractor provided with an ultrasonic system to assist
585 the extraction (Riera et al., 2004).

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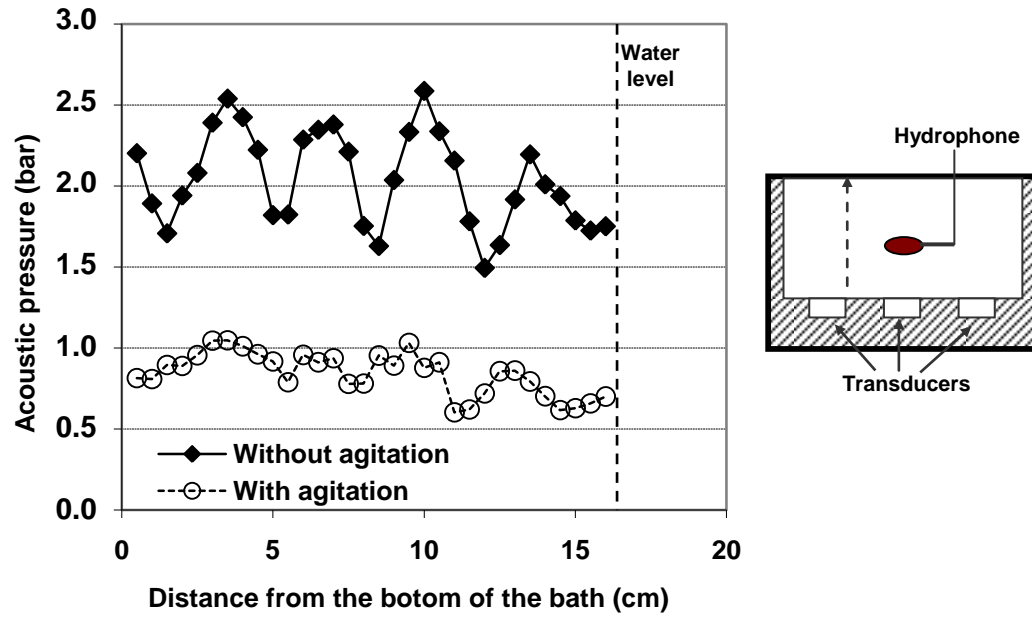
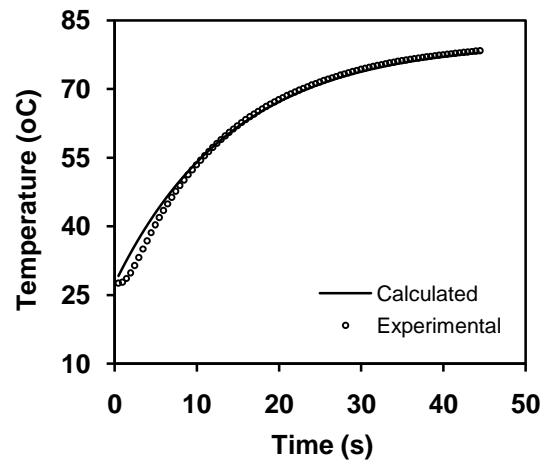
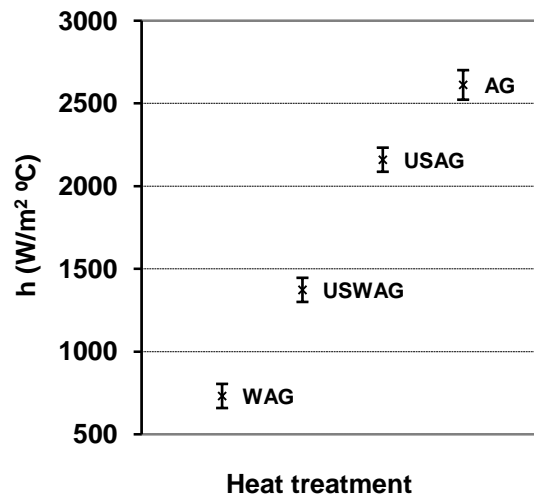


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A



B

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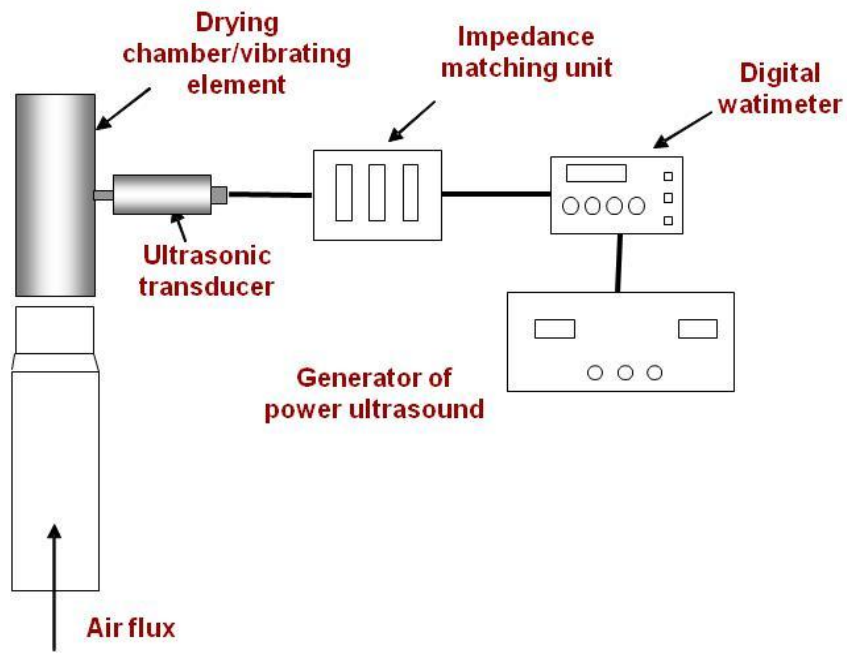


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Figure 4

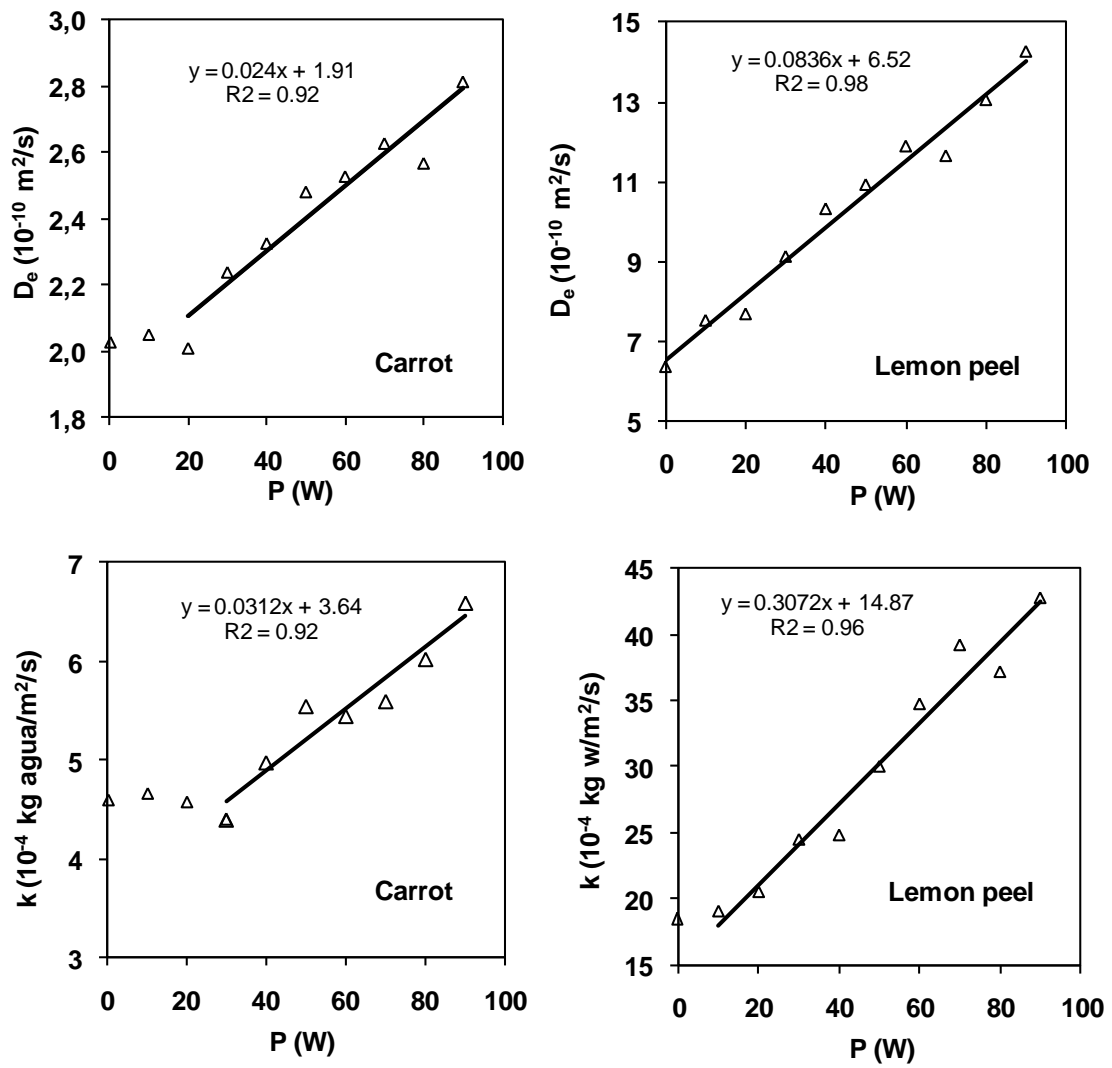


Figure 4. Identified effective diffusivity (D_e) and mass transfer coefficient (k) for the drying of carrot and lemon peel with ultrasound application at different acoustic powers. Air temperature of 40 °C and air velocity 1 m/s (García-Pérez et al. 2009).

Figure 5

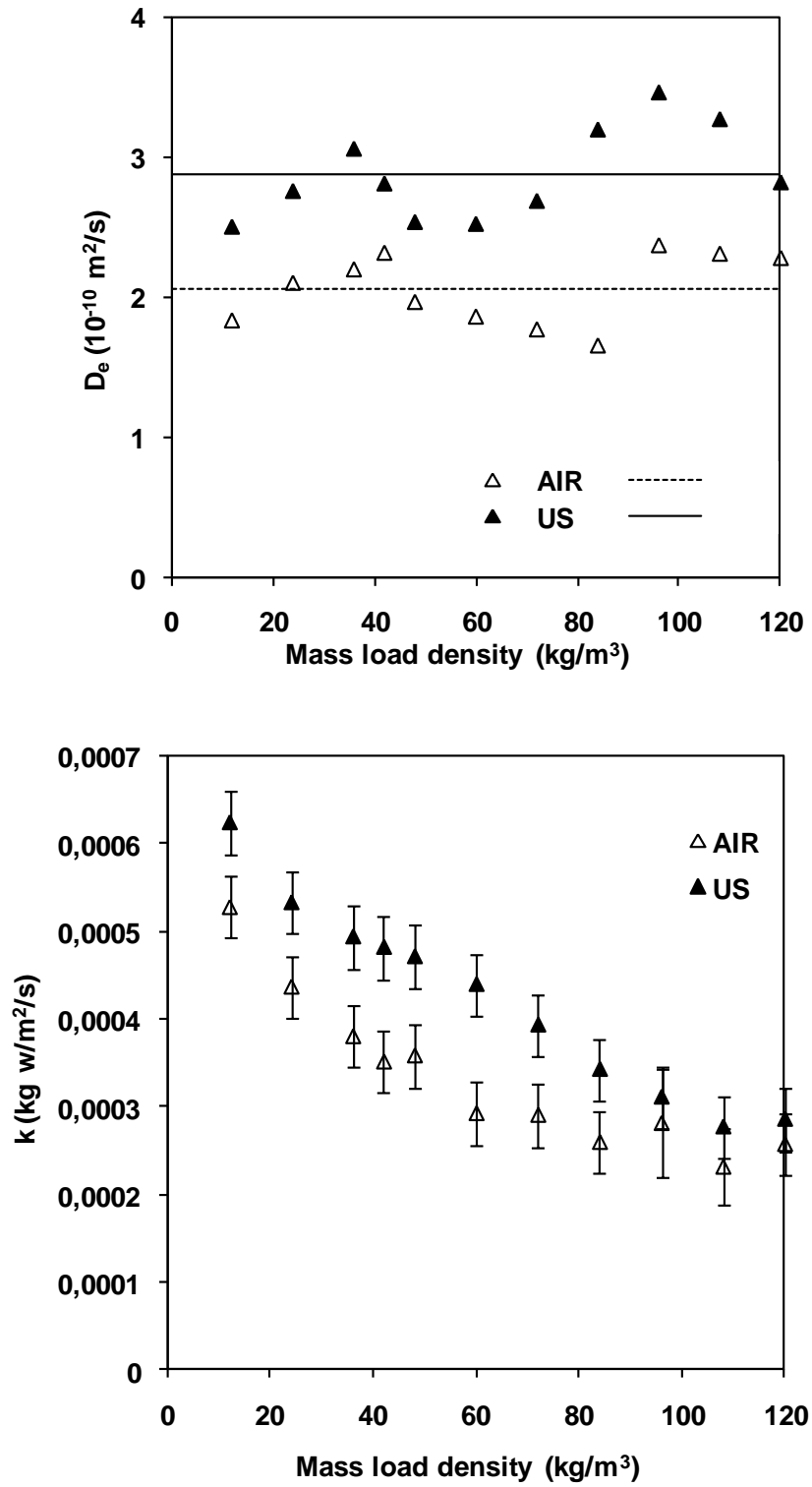


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Figure 6

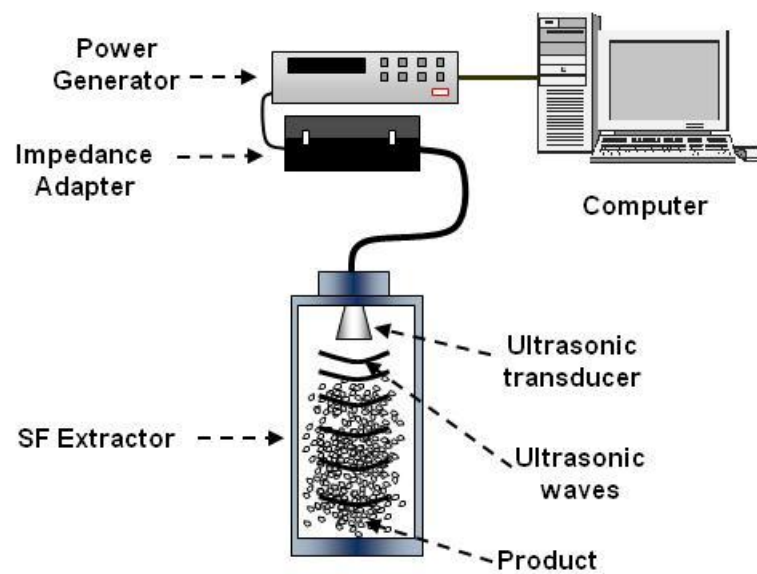


Figure 6. Scheme of the supercritical fluid extractor provided with an ultrasonic system to assist the extraction (Riera et al., 2004).

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	Percentage of the applied electric power				
	20	40	60	80	100
Brine	0.63 ± 0.07	0.68 ± 0.02	0.73 ± 0.05	0.88 ± 0.01	0.92 ± 0.09
Sucrose solution	0.56 ± 0.05	0.60 ± 0.04	0.66 ± 0.03	0.77 ± 0.06	0.86 ± 0.05