EXTENDED ABSTRACT PhD THESIS

"CONTRIBUTION TO THE KNOWLEDGE ON THE USE OF POWER ULTRASOUND IN CONVECTIVE DRYING OF FOODS"

Author: Dr. Jose V. Garcia-Perez (Start Date: January 2003, Defense Date: May 2007)

Advisors: Antonio Mulet and Juan A. Carcel

Universidad Politécnica de Valencia, Spain.

1. SUMMARY

The use of non conventional energy sources during convective drying to increase the drying rate without affecting quality attributes is highly relevant due to the public concern on reducing energy consumption and consuming high quality products. Power ultrasound application could constitute a way to enhance food drying in order to improve not only mass transfer but also product quality, since it does not significantly heat the material. The main aim of this PhD was to evaluate the application potential of power ultrasound on convective drying. In order to reach this goal, the influence of the main process variables was addressed.

A vibrating cylinder, constituting not only the drying chamber but also the ultrasonic radiating element, was developed and installed in a conventional convective drier. Convective drying kinetics were carried out with (21.8 kHz, US experiments) and without ultrasound application (NUS experiments) at different air velocities, temperatures, ultrasonic power applied, mass load densities and using different products (carrot, lemon peel, persimmon and apricot). Modelling was a fundamental tool to quantify and clarify the influence of power ultrasound on mass transfer phenomena involved on convective drying.

The results showed that power ultrasound application involved a significant improvement of drying rate, showing drying time reductions over 50%. From modelling, it was established that effective moisture diffusivities were increased up to 55% by ultrasound application. The influence of power ultrasound depended on the magnitude of the process variables. Thereby, the effect of power ultrasound was more intense at low air velocities and temperatures, high ultrasonic powers applied and porous products. Therefore, the application of power ultrasound on convective drying processes may result highly relevant.

2. INTRODUCTION (State of the art, relevance, main goal and literature)

Convective drying of foods presents some limitations which make it difficult to apply on specific fields (Lewicki, 2006). Among other things, the low drying rate and product quality loss must be considered besides the large energy needs (Kudra and Mujumdar, 2002). Some of these limitations may be overcome by combining other technologies (Mujumdar, 2006), which may be used as additional energy sources during drying. Power ultrasounds are considered as an appropriate technology since they may affect drying without significantly heating the material (Gallego-Juarez et al., 1999). This fact may contribute to its application in the drying of heat sensitive materials or in drying processes carried out at low temperatures, such as atmospheric freeze drying.

Power ultrasounds have been applied to affect mass transfer processes in solid-liquid treatments, like meat (Mulet et al., 2003) and cheese brining (Sanchez et al., 1999), the osmotic dehydration of fruits (Mulet et al., 2003) and several extraction processes (Riera et al., 2004). Nevertheless, applications on solid-gas systems are much less frequent due to some technical difficulties, which prevent this technology from being fully developed. Among other things, the high impedance mismatch between the application systems and air, which makes the acoustic wave transmission difficult, and the high acoustic energy absorption of the air must be considered. These limitations may be overcome by adequately designing the ultrasonic application system (Gallego-Juarez et al., 1999).

The mass transfer process that takes place during convective drying may be influenced by a series of effects associated to power ultrasound application (Gallego-Juarez et al., 1999; Mulet et al., 2003). On one hand, the external resistance to mass transfer may be affected by pressure variations, oscillating velocities and microstreaming at the solid-gas interfaces thus reducing the boundary layer thickness and therefore improving water transfer from solid surface to air medium. On the other hand, internal resistance may be reduced by alternating expansion and compression cycles produced by ultrasound in the material (a phenomenon known as the “sponge effect”) and also through some effects on the interfaces of intercellular spaces, or even by cavitation which may contribute to removing the strongest attached moisture to solid matrix.

The main aim of this work was to determine the effect of applying power ultrasound on convective drying processes, establishing the influence of the main process variables on different products.

LITERATURE

Gallego-Juárez, J.A., Rodríguez-Corral, G., Gálvez-Moraleda, J.C., Yang, T.S. (1999). A new high intensity ultrasonic technology for food dehydration. Drying Technology 17: 597-608.

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3. TECHNOLOGICAL INNOVATION. DEVELOPMENT OF AN ULTRASONICALLY ASSISTED CONVECTIVE DRIER

In order to reach this objective, a new ultrasonic system was designed to attain a good transmission of acoustic energy to the gas medium. The design was carried out by considering the walls of the drying chamber as the device for radiating the acoustic energy, and as a consequence the drying chamber would constitute the vibrating element transmitting the acoustic energy to the particles. The design was made through modelling, considering a finite element model in 2 dimensions using the ANSYS code. As a result, an ultrasonic system consisting of an aluminium cylinder (external diameter 120 mm, thickness 10 mm and height 310 mm) driven by a piezoelectric composite transducer was developed. The driving transducer consists of an extensional piezoelectric sandwich element together with a mechanical amplifier. The ultrasonic system was characterized before being installed on the convective drier, and for that purpose not only the electric properties were measured using an impedance analyzer but also the response of the system when applying an electric power of 90 W. Thereby, the ultrasonic parameters measured in the ultrasonic signal were: frequency 21.8 kHz, voltage 60 V, intensity 1.55 A, phase 4º and impedance 329 Ω. The acoustic field generated inside the cylinder produced an average sound pressure level of about 154.3 dB when the electrical power applied to the transducer was 75 W. This figure was very close to that estimated from modelling using the finite element method (156.3 dB).

A laboratory scale convective drier was modified to install the different elements of the ultrasonic system. The drier modifications were carried out in such a way as to provide good working conditions for the vibrating cylinder and also to maintain the automatic weighing system on the drier.

4. MAIN RESULTS AND DISCUSSION

Drying kinetics of several foodstuffs were carried out: carrot, apricot, persimmon and lemon peel in order to identify the influence of the main process variables. In all the cases, sorption isotherms were obtained from literature except for lemon peel, since no references were found for this product. Thus, the sorption isotherms of lemon peel were determined at different temperatures (20, 30, 40 and 50 ºC) using electric hygrometers.

The GAB model was rated as the best for describing the experimental sorption data when considering the influence of temperature. The isosteric heats of sorption were determined from the Clausius-Clapeyron equation using differential and integral identification methods and also from Riedel equation. The isosteric heats of sorption provided by both methods were very similar.

The influence of power ultrasound on mass transfer process that occurs during convective drying, as well as, the effect of the main process variables on the ultrasonic application are summarized as follows. In order to quantify the influence of ultrasound, not only mechanistic models with different degree of complexity but also empirical models were used.

4.1. Air velocity and material's structure

No significant effect of power ultrasound application (75W, 21.7 kHz) was observed on fluidized bed drying kinetics of carrot cubes (side 8.5 mm) and eights parts of apricots. Experiments were carried out at different temperatures (30, 35, 40, 45, 50, 55 and 60 ºC) and air velocities between 10 and 14 m/s. Activation energy figures were similar to others found in the literature for these products. The results from these experiments suggested an influence of air velocity on the ultrasonic field.

Further experiments showed that as the air velocity increased, there was a reduction in the average sound pressure level, although, it seems to remain constant from an air velocity figure of about 8 m/s onwards. Therefore, the air velocity increase provided lower acoustic energy levels available for the particles in the drying chamber. As a consequence, it is of interest not to use velocities higher than a given threshold in a particular application.

Drying kinetics of different products (carrot, persimmon and lemon peel) and geometries (cubes, cylinders and disks) were carried out at air velocities of between 0.5 and 14 m/s. Air velocity affected the non ultrasonic (NUS) drying kinetics of the different products up to a value of about 5 m/s. This threshold was established from the effective moisture diffusivities (D_e) identified with a diffusion model considering external resistance to mass transfer as negligible (NER model). Power ultrasound application (75 W, 21.7 kHz) only increased the effective diffusivity values identified with the NER model on the drying kinetics of carrot and persimmon for experiments carried out at air velocities lower than 6 m/s. As a consequence, the influence of power ultrasound was negligible at high air flow rates (low acoustic energy levels considered). However, power ultrasound application on lemon peel drying significantly increased ($p<0.05$) effective diffusivities at all the air velocities considered. A possible explanation of this finding is that lemon peel behaves differently compared to carrot and persimmon due to its structure. Acoustic effects on lemon peel were greater, as it is considered to be a more porous product than carrot or persimmon.

Porosity may be considered as one of the most important structural variables for determining the acoustic effectiveness in foodstuffs. High porosity products may be considered more prone to alternating compression and expansion cycles produced by ultrasonic waves, improving water movements in its large pores and intercellular spaces. Small intercellular spaces are also found in low porosity products, that means a high internal resistance to mass transfer. Thus, high acoustic energy levels are required to affect mass transfer in low porosity products.

The influence of porosity may also be explained considering a greater acoustic energy absorption in high porosity products. As a consequence, the internal energy available in the particles would increase, leading to more intense compressions and expansions (sponge effect), which could improve water removal and therefore, reduce internal resistance. Furthermore, the acoustic effects on the solid-gas interfaces of intercellular spaces could increase in high porosity products due to a larger porous net. Indeed, this phenomenon also contributes to reduce internal resistance to mass transfer.

Diffusion models that do not consider the external resistance to mass transfer (NER) presented a poor fit to the experimental data in experiments carried out at low air velocities but a good fit at high air velocities. These results will indicate that when external resistance decreases linked to the increase of air velocity, NER model would apply. Therefore, diffusion models considering external resistance (ER), which differ according to geometry, were considered on experiments carried out at low air velocities. ER models were solved using an implicit finite difference method using the programming language available on Matlab. ER models provided percentages of explained variance higher than 99 % and mean relative errors lower than 10 % in all cases.

In persimmon drying experiments carried out at air velocities lower than 6 m/s, power ultrasound application (75 W, 21.7 kHz) significantly increased ($p<0.05$) the effective moisture diffusivity (D_e) and the mass transfer coefficient (k) values identified with the ER model. Therefore, both external and internal resistance to mass transfer were significantly affected by power ultrasound application when low air velocities were used.

4.2. Air temperature

The influence of air temperature on power ultrasound assisted convective drying was addressed from NUS (without ultrasound) and US (ultrasound, 75 W, 21.7 kHz) drying experiments of carrot cubes (side 8.5 mm) carried out at 1 m/s and at several air temperatures: 30, 40, 50, 60 and 70 ºC. D_e and k figures identified from US experiments were only significantly ($p<0.05$) higher than those identified on NUS experiments when air temperature was lower than 60 ºC. The influence of power ultrasound application decreased with the increase of air temperature and it was almost negligible at 70 ºC. Effective moisture diffusivities identified from US experiments at the different temperatures showed a poor fit when considering an Arrhenius equation, due to the cross effect of temperature and ultrasound application. Figures identified at high temperatures (60 and 70 ºC) departed from the tendency shown by D_e figures identified at low temperatures (30, 40 and 50 ºC). Due to the increased thermal energy available for water removal at higher temperatures the input from ultrasounds becomes comparatively almost negligible.

4.3. Mass load

The effect of mass load used in the experiments was also considered. In order to clarify its influence, NUS and US (75 W, 21.7 kHz) drying experiments of carrot cubes (side 8.5 mm) were carried out at several mass load densities: 12, 24, 36, 42, 48, 60, 72, 84, 96, 108 and 120 kg/m^3, at 1 m/s and at 40 ºC. There was a significant ($p<0.05$) influence of mass load on drying kinetics. The drying rate decreased as the mass load placed in the drying chamber increased. The results provided by the ER model showed that mass load density did not affect the value identified for D_e. Mass load density only affected the mass transfer coefficient (k). The increase on the number of particles placed on the drying chamber trays may disrupt the air flow around the particles and create channelling. This fact increases external resistance.

The influence of power ultrasound was observed in the whole range of mass load density tested increasing both D_e and k figures. Nevertheless, the effect was not significant ($p<0.05$) at mass load densities higher than 90 kg/m^3. This fact may be explained considering the decrease of the amount of energy available per unit mass as the load increases and also due to the screen effect of particles due to energy absorption.

4.5. Ultrasonic power applied

An important variable to take into account is the ultrasonic power level applied. Drying experiments of carrot cubes (side 8.5 mm) and lemon peel slabs (thickness 7 mm) were carried out at 1 m/s and 40 ºC at different ultrasonic powers: 0, 10, 20, 30, 40, 50, 60, 70, 80 and 90 W. A significant ($p<0.05$) influence of this variable on the drying kinetics was observed. In the case of lemon peel, a linear correlation was found between both D_e and k with the ultrasonic power applied, this relationship was valid for all the range tested. Nevertheless in carrot drying, the ultrasonic effects were negligible below ultrasonic power figures of around 20-30 W. Above this threshold, the linear relationship between both D_e and k with the power ultrasonic was also observed. Furthermore, the slopes found in the linear relationships were 10 times lower in the case of carrot regarding those found on lemon peel experiments. As a consequence, the ultrasonic effects were more intense on lemon peel (porosity 0.4) than in carrot (porosity 0.04), which is considered a low porosity product. Therefore, the influence of raw material structure on the ultrasonic effects observed during convective drying is confirmed and should be taken into consideration when planning/designing drying operations.

4.6. Empirical models

To test if the conclusions reached demonstrated a bias in the model, the empirical model of Weibull was used in a complementary way to NER and ER diffusion models to describe the different kinds of drying experiments. The Weibull model adequately described the drying kinetics of the different products, providing percentages of explained variance similar to those found by the RE model and much higher than the NER model at low air velocities. The Weibull parameters (α and β) convey similar information about the influence of power ultrasound on convective drying to that obtained through diffusion models. This will indicate that conclusions reached were not biased by the models applied.

5. POTENTIAL APPLICATIONS. ULTRASONICALLY ASSISTED ATMOSPHERIC FREEZE DRYING

Atmospheric freeze drying technology was addressed during PhD in order to consider the use of power ultrasound in this process. Atmospheric freeze drying presents low drying rates due to the use of air temperatures below freezing point. Power ultrasound application may be considered interesting in these conditions, since it could increase the drying rate without significantly heating the material. Thus, addressing ultrasonic application in this process may be considered as a matter of relevant research. Before addressing ultrasonic application, the influence of atmospheric freeze drying on the quality parameters of a highly valuable product, like cod fish, was considered. For that purpose, the PhD candidate stayed for 3 months (under Marie Curie Program-Training Sites in EcoEff Driers) in the Norwegian University of Science and Technology/SINTEF Energy under the supervision of Dr. Alves-Filho and Prof. T.M. Eikevik. The stay in the NTNU also allowed obtaining the European PhD label.

During the stay in the NTNU, drying kinetics of granulated and cubic (side 5 mm) samples of cod fish were carried out at different temperatures (-10, -5, 0, 15 and 30 ºC) using a heat pump drier. Experiments were also conducted by combining temperatures. Temperatures below freezing point (-10 or -5 ºC) were used until moisture contents of close to 0.4 kg water/kg product, while a high temperature (30 ºC) was applied in the final drying stage. Drying kinetics of cod fish cubes were fitted using diffusion (NER) and Weibull models. Different activation energy figures were assessed from the effective moisture diffusivities identified for experiments carried out below freezing point (-10 and -5 ºC, 71.1 kJ/mol) and at high temperatures (15 and 30 ºC, 30.7 kJ/mol). Higher quality parameters were found for samples dried below freezing point, these samples presented higher brightness, lower shrinkage, lower bulk density and higher rehydration ability than samples dried using hot air (15 and 30 ºC). Samples dried at 0ºC, on the other hand, presented intermediate quality parameters. The combination of temperatures (-10/30ºC or -5/30 ºC) significantly increased the drying rate and provided similar quality parameters in the samples compared to those dried only under atmospheric freeze drying conditions.

Power ultrasound application would increase mass transfer rate in atmospheric freeze drying processes without significantly heating the material, and therefore without affecting product quality parameters. Based on ultrasonic experiments an effective moisture diffusivity increase about 55 % would be expected by ultrasound application, similar to that obtained on experiments carried out in this work. As a consequence, the drying time to reach a moisture content of 0.15 kg water/kg product would be reduced by 45000 s (12.5 hours) in experiments carried out at -10 ºC.

The ultrasonically atmospheric freeze drying could constitute a potential technology for water removal not only in the food industry but also in other relevant sectors, such as pharmaceutical and biotechnology industries. Actually, these industries require non thermal dehydration techniques due to the products' bio-activity is frequently affected by heating. Therefore, ultrasonically atmospheric freeze drying technology would provide products with high quality attributes at lower energy costs with regards to traditional freeze drying.

Due to the potential use of power ultrasound in atmospheric freeze drying and the full development of this application, a research project has been recently funded by the Spanish Ministry of Science and Innovation, the award's candidate being part of the research team. The project involves the development of an ultrasonically assisted atmospheric freeze drier prototype, and the study of the influence of power ultrasound on mass transfer. The participation of the Spanish Association of Food Industry as project's partner evidences the potential application of this technology at industrial scale. Some details of this project are:

Title: Study of power ultrasound effects on mass transfer processes. Improvement of atmospheric freeze drying
Research team of Polytechnic University of Valencia: A. Mulet, J.A. Cárcel, **J.V. Garcia-Perez**
Partners: Polytechnic University of Valencia, Institute of Acoustics, University of Balearic Islands and Spanish Association of Food Industry.
Total budget: 500.000 €

6. PUBLICATIONS

The results obtained linked to the PhD work have been spread in journals' papers and International Congresses, in addition, the results have supported partially three book chapters. The number and quality of publications determines the degree of diffusion and relevance of this PhD Thesis.

Papers listed in the ISI Web of Knowledge:

- **García-Pérez, J.V.**, Cárcel, J.A., De la Fuente, S., Riera, E. (2006). Ultrasonic drying of foodstuff in a fluidized bed. Parametric study. Ultrasonics 44: e539-e543.
- **García-Pérez, J.V.**, Rosselló, C., Cárcel, J.A., De la Fuente, S., Mulet, A. (2006). Effect of air temperature on convective drying assisted by high power ultrasound. Defect and Diffussion Forum 258-260: 563-574.
- **García Pérez, J.V.**, Cárcel, J.A., Benedito, J., Mulet, A. (2007). Power ultrasound mass transfer enhancement in food drying. Food and Bioproducts Processing 85: 247-254.
- Cárcel, J. A., **García-Pérez, J.V.**, Riera, E., Mulet, A. (2007). Influence of high intensity ultrasound on the drying kinetics of persimmon. Drying Technology 25:185-193.
- **García-Pérez, J.V.**, Cárcel, J. V., Clemente, G., and Mulet, A. (2008). Water sorption isotherms of lemon peel at different temperatures and isosteric heats. LWT- Food Science and Technology 41: 18-25.
- **García-Pérez, J.V.**, Cárcel, J.A., Benedito, J., Riera, E., Mulet, A. (2008). Drying of a low porosity product (carrot) as affected by power ultrasound. Defect and Diffusion Forum 273-276: 764-769.
- **García-Pérez, J.V.**, Cárcel, J.A., Riera, E., Mulet, A. (2009). Influence of the applied acoustic energy on the drying of carrots and lemon peel. Drying Technology 27: 281-287.

- Cárcel, J.A., **García-Pérez, J.V.**, Riera, E., Mulet, A. (2010). Influence of mass load density on ultrasonically assisted forced air drying of carrot cubes. Drying Technology, in press.

Book chapters:

- Mulet, A., **García-Pérez, J.V.**, Cárcel, J.A., Riera, E. 2010. Ultrasound-assisted hot air drying of foods. In Ultrasound technologies for food and bioprocessing. Ed. Gustavo G.V. Barbosa-Canovas y Hao-Fengh, Springer, in press.
- Mulet, A., Cárcel, J.A., Sanjuán, N., **García-Pérez, J.V.** 2010. Food dehydration under forced convection conditions. In Recent Progresses in Chemical Engineering. Ed. Joao Delgado, Publishing House Studium Press LLC, in press.
- Riera, E., **García-Pérez, J.V.**, Acosta, V.M., Carcel, J.A., Gallego-Juárez, J.A. 2010. A computational study of ultrasound-assisted drying of food materials. In Multiphysics Simulation of Emerging Food Processing Technologies. Ed. Kai Knoerzer, Pablo Juliano, Peter Roupas and Cornelis Versteeg, IFT Press, in press.

Contributions to Congresses:

- *IDS-International Drying Symposium:*

  García-Pérez, J.V., Bon, J., Sanjuán, N., De la Fuente, S., Mulet, A. (2006). Mass loading effect on ultrasonic drying of carrots. IDS2006. Poster.

  Cárcel, J. A., Clemente, G., **García-Pérez, J.V.**, Ricarte, B., Mulet, A. (2006). Study of the influence of air velocity on the drying of persimmon using a diffusion model. IDS2006. Poster.

  García-Pérez, J.V., Cárcel, J.A., Bon, J., Simal, S., Mulet, A. (2006). Isosteric heats of sorption of lemon peel. IDS2006. Poster.

  García-Pérez, J.V., Carcel, J.A., García-Alvarado, M.A., Riera, E., Mulet, A. (2008). Effect of power ultrasound on second law thermal efficiency of food convective drying. IDS2008. Poster.
- *European Drying Conference-AFSIA/EFCE:*

  García-Pérez, J.V., De la Fuente, S., Riera, E., Simal, S., Mulet, A. (2005). Hot air drying assisted by power ultrasound: Effect of air flow rate. AFSIA2005. Poster and Presentation.

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  García-Pérez, J.V., Cárcel, J.A., Rosselló, C., De la Fuente, S., Mulet, A. (2007). Drying of porous materials as affected by power ultrasound. AFSIA2007. Poster and Presentation.
- *Nordic Drying Conference:*

  García-Pérez, J.V., Alves-Filho, O., Eikevik, T.M., Strommen, I., Mulet, A. (2005). Heat pump fluidized bed drying of cod fish. NDC2005. Presentation.
- *International Conference on Drying and Hygro-Thermal Processing:*

  García-Pérez, J.V., Cárcel, J.A., Riera, E., Gallego-Juárez, J.A., Mulet, A. (2005). Power ultrasonic effect on the drying of carrots. DHTP2005. Poster and Presentation.

  Carcel, J.A., Bon, J., **García-Pérez, J.V.**, Gonzalez, R., Mulet,A. (2005). Influence of air velocity on the drying kinetics of persimmon. DHTP2005. Poster and Presentation.
- *World Congress on Ultrasonics/Ultrasonics International:*

García-Pérez, J.V., Cárcel, J.A., De la Fuente, S., Riera, E. (2005). Ultrasonic drying of foodstuff in a fluidized bed. WCU/UI2005. Presentation.

- *International Congress on Acoustics:*

  García-Pérez, J.V., Cárcel, J.A., Benedito, J., Riera, E., Mulet, A. (2007). Influence of process variables on hot air drying assisted by power ultrasound. ICA2007. Presentation.

- *IFT-Annual Meeting Institute of Food Technologists:*

  García-Pérez, J.V., Cárcel, J.A., Riera, E., Gallego-Juárez, J.A., Mulet, A. (2009). A computational study of ultrasound-assisted drying of food. IFT2009. Presentation.

- *International Conference on Diffusion in Solids and Liquids:*

  García-Pérez, J.V., Rosselló, C., Cárcel, J.A., De la Fuente, S., Mulet, A. (2006). Effect of air temperature on convective drying assisted by high power ultrasound. Presentation. DSL2006.

  García-Pérez, J.V., Cárcel, J.A., Benedito, J., Riera, E., Mulet, A. (2007). Drying of low porosity products as affected by power ultrasound. DSL2007.

- *Congreso Iberoamericano de Ingeniería de Alimentos:*

  García-Pérez, J. V., Cárcel, J. A., Riera, E., Mulet, A. (2007). Aplicación de ultrasonidos de alta intensidad en el secado convectivo de alimentos. CIBIA2007. Conference.

- *Technical and Business Meeting of EFCE Working Party on Drying:*

  Mulet, A., **García-Pérez, J.V.**, Cárcel, J.A., Sanjuán, N., Benedito, J. (2008). Application of power ultrasound on drying. Poster and Presentation.

7. AWARDS

The PhD thesis has been already awarded with the “Excellence award of Doctoral Thesis” given by the Polytechnic University of Valencia, Academic Curse 2006/2007. This award supports the nomination of this work for the EFCE Excellence Award in Drying 2010.