Available online at www.sciencedirect.com

ScienceDirect

Physics Procedia 3 (2010) 141–146

Physics Procedia

www.elsevier.com/locate/procedia

International Congress on Ultrasonics, Universidad de Santiago de Chile, January 2009

High-power ultrasonic system for the enhancement of mass transfer in supercritical CO_2 extraction processes

Enrique Riera[a]*, Alfonso Blanco[b], José García[c], José Benedito[d], Antonio Mulet[c], Juan A. Gallego-Juárez[a], Miguel Blasco[c]

[a] *Grupo de Ultrasonidos de Potencia, Dpto. Señales, Sistemas y Tecnologías Ultrasónicas, Instituto de Acústica, CSIC, Serrano 144, E28006-Madrid, Spain*

[b] *Instituto de Física Aplicada, CSIC, Serrano 144, E28006-Madrid, Spain*

[c] *Centro Tecnologico AINIA, Benjamin Franklin 5-11, E46980-Paterna Valencia, Spain*

[d] *Grupo de Análisis y Simulación de Procesos Agroalimentarios; Dpto. Tecnología de Alimentos, Universidad Politécnica de Valencia, Camino de Vera s/n, E46022-Valencia, Spain*

Abstract

Oil is an important component of almonds and other vegetable substrates that can show an influence on human health. In this work the development and validation of an innovative, robust, stable, reliable and efficient ultrasonic system at pilot scale to assist supercritical CO_2 extraction of oils from different substrates is presented. In the extraction procedure ultrasonic energy represents an efficient way of producing deep agitation enhancing mass transfer processes because of some mechanisms (radiation pressure, streaming, agitation, high amplitude vibrations, etc.). A previous work to this research pointed out the feasibility of integrating an ultrasonic field inside a supercritical extractor without losing a significant volume fraction. This pioneer method enabled to accelerate mass transfer and then, improving supercritical extraction times. To commercially develop the new procedure fulfilling industrial requirements, a new configuration device has been designed, implemented, tested and successfully validated for supercritical fluid extraction of oil from different vegetable substrates

Keywords: Ultrasonic processes; Power ultrasound; Mass transport; Supercritical Fluid extraction; Carbon dioxide

1. Introduction

At present, supercritical CO2 is considered in the food sector as an excellent solvent in the product extraction from vegetables. Extraction with supercritical carbon dioxide is also considered an environmentally friendly technology which has gained wide acceptance as an alternative to conventional solvent extraction because of its important advantages (non-toxic, recyclable, cheap, relatively inert and non-flammable). Nevertheless, the economics of supercritical fluid extraction (SFE) is affected by the slow kinetics of the process. Since high pressures

* Corresponding author. Tel.: +34-91-561-8806; fax: +34-91-411-7651.
E-mail address: eriera@ia.cetef.csic.es.

doi:10.1016/j.phpro.2010.01.020

are normally used in SFE, mechanical stirring is difficult to be applied. The application of new techniques such as the use of high-power ultrasound (HPU) assisting this process has proved important benefits as a consequence of the mechanical effects produced in the supercritical environment through the high amplitude vibrations, radiation pressure, streaming, agitation, etc.) [1-5].

Nowadays, high-intensity ultrasound is regarded as an emergent technology in food, chemical and pharmaceutical industries [6, 7]. In particular, ultrasonic energy represents a clean way to accelerate and improve mass transfer processes. In addition, the use of energy would permit to obtain economically competitive high quality food extracts. Previous studies indicate the improvement of the supercritical CO_2 extraction of almond oil by using HPU at laboratory scale [8]. Specifically, the application of HPU to assist extraction process produced a relevant increase in the final yield of the oil together with a noticeable reduction of the energy consumption through the increase of the extraction rate.

This paper deals with the implementation and validation at pilot scale of an innovative ultrasonic system to assist supercritical CO_2 extraction which aims to be a robust, stable, reliable and efficient solution to be applied at industrial scale. The ultrasonic device was designed and built to work at 19 kHz and to overcome the fluctuations of the specific acoustic impedance of the fluid medium during the extraction process under the operational conditions (high pressure ≤ 400bar, temperatures up to 70°C, mass flow up to 20kg/h and density of the CO_2 ≤ 900g/cm^3). The ultrasonic applied power was maintained constant during the extraction trials of oils from different substrates. A specific hardware and software has been developed, tested and validated to control and monitoring the parameters involved in the supercritical extraction process assisted by power ultrasound.

2. Experimental

2.1. Ultrasound-assisted supercritical fluid extraction (USFE) system

As showed in [9] the USFE system is made up of two units: a) SFE unit and b) HPU unit. The SFE pilot plant (AINIA, Valencia) was built to withstand high pressures up to 350 bar and temperatures up to 80ºC (see Fig.1 and Fig.2). The major components of SFE include a diaphragm pump, a high pressure extraction vessel with 5L capacity, two separation units (a cyclone and a decanter) and a set of sensors to monitor and control temperature, pressure and fluid flow rate. High purity CO_2 (99.9%) was used as a solvent in the extractor under supercritical conditions (P_c=73.9 bar; T_c = 31.1ºC; D_c = 0.468 g/mL) [10]. CO_2 was used because is the most commonly used solvent for the extraction of oils from vegetables in supercritical processes due to its excellent behaviour and low cost.

Fig.1 SFE pilot plant assisted by power ultrasound

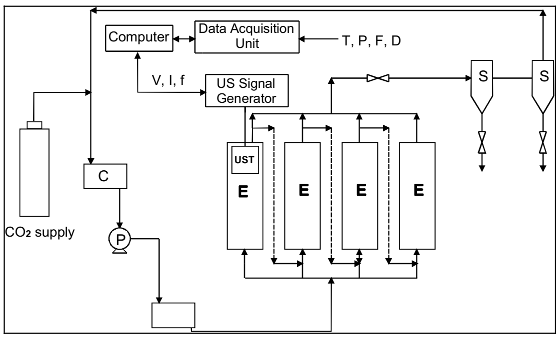

Fig.2 Scheme of the SFE pilot plant assisted by power ultrasound

In the previous USFE device [2] an operator was in charge of the control of the operational conditions in the SFE as well as the control of the ultrasonic system. Although promising results were obtained in the almond-oil extraction tests, carried out at 280 bar, 55ºC and a flow rate of 20 kg/h with an ultrasonic transducer operating at a frequency of 18 kHz and a power of 50W (faster kinetic curves ~30% and higher extraction yields ~20%), some instabilities in its behaviour were detected. Those instabilities were caused by the changes observed in the acoustic impedance of the medium under supercritical conditions during the extraction process. In other words, the ultrasonic system exhibited difficulties concerning stability and that made it unable for industrial use. In parallel, resonance modes of the metallic basket placed inside the extractor (where the substrate is deposited) were detected in the first prototype giving rise to modal interactions that disturb operation. In the new system such interactions have been eliminated by separating both basket and transducer resonant frequencies up to a value of about 1 kHz. To achieve it, the resonant frequency and the shape of the new transducer was established by finite element methods (FEM) In this work special attention has been given to the transducer characteristics (impedance, phase and power spectrum of the current) as a function of extraction parameters (pressure, temperature, flow rate, density), power applied and time during the extraction operation.

The ultrasonic system presented here is a new version of the previous one and can be considered as the second step to scale it up for larger operation. In fact, the new device implemented definitive advantages related to the automatic control required for industrial applications. The present ultrasonic system basically consists of: a) the ultrasonic transducer to work at 19 kHz with a maximum power of about 110W; b) a dynamic resonance control unit (controller); c) a broadband power amplifier, d) an impedance matching unit, e) a specific developed software to monitor and control the parameters of the power transducer (voltage V, current I, phase φ, impedance Z, power P and frequency f) and those of the supercritical fluid (pressure P, temperature T, flow rate F and density D), and f) a computer with a data acquisition hardware. To achieve automatic control, the new system, also allows the power characterization of the transducer during the extraction process in real time. To this purpose, a virtual high-power impedance analyzer for continuous operation was developed with LabView code, tested and validated experimentally [9]. In this way, the transducer behaviour during the extraction process and the enhancement by ultrasound on the kinetic curves and the oil extraction yields of two different products, have been analyzed.

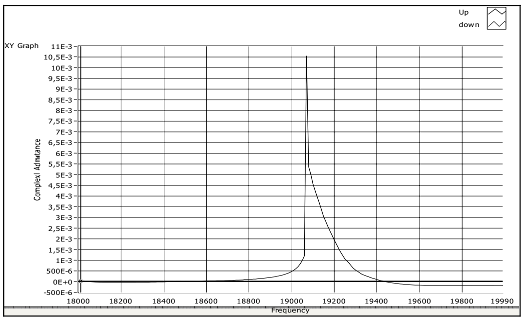

Fig.3 Conductance of the transducer versus frequency at 100W

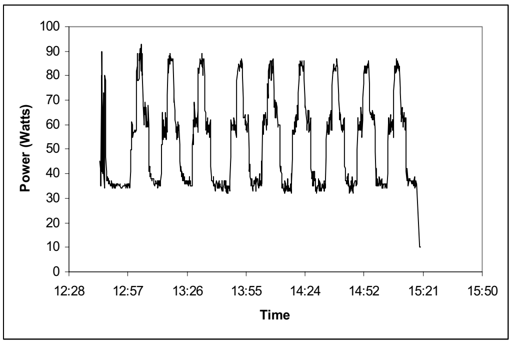

Fig.4 Example of power modulation applied to the transducer

2.2. Materials and Methods

Two examples of the potential of HPU in SFE processes are presented in this work: a) grounded almonds sieved at 3-4 mm in size (55% oil content, wet basis), and b) a second grounded vegetable product "cocoa cake" with similar particle size. In both cases, an amount of about 1500 g, were deposited in the basket placed in the extractor of 5L capacity. The selection of the particle size of the samples (3-4 mm) was done based on our previous results with grounded almonds which confirm that small particulate size favours the ultrasonic action. In addition, experimental trials were carried out with a second substrate cocoa cake. The time for each trial was about 3.5 to 4 hours. The power output applied to the ultrasonic transducer was fixed and kept constant at 85W in all the experiments here presented.

3. Results and Discussion

The ultrasonic transducer was placed inside the extractor inserted on the upper part of the vessel. At low power, an impedance analyzer was used to measure the admittance response of the transducer in the frequency range (19 – 20 kHz). Only one vibration mode was detected in air at 19228 Hz with an impedance of 90 Ω. The same mode was detected at high excitation (see Fig. 3) but at 19.1 kHz. The transducer has an estimated power capacity of 110W.

3.1. Stability of the prototype

The power behaviour of the new transducer driven at high-excitation level was studied in air and inside the extractor during the extraction process with supercritical CO_2. First, the stability of the prototype was tested and validated in air at 100W during a long time. The new transducer showed high-stability and good performance during 8 hours of continuous operation at its maximum power capacity. Such experiment was repeated several times up to reach a total number of 50 hours following the high-power characterization procedure described in [11]. No change was detected in its behaviour during the trials. Therefore, the power characterization of this transducer validates it for SFE. In addition, other kind of driving signals, were applied to study the transducer behaviour in air. One example consisted in applying a power modulation between 35W and 90W to analyze the transducer response. Fig.4 shows the response that can be considered very reliable.

The transducer was also characterized at high level excitation inside the extractor of the SFE unit in order to study the effect of the high-pressure conditions on its behaviour. Its impedance increases from 90Ω up to 260Ω, and the frequency decreases from 19.2 kHz up to 18.9 kHz when the value of the pressure varies from 200 bar up to 320 bar. In Fig.5 it is plotted the evolution of the power applied to the transducer versus time along one extraction trial carried out at 85W. It is clear from the picture that the response of the ultrasonic device is quite stable also in this case.

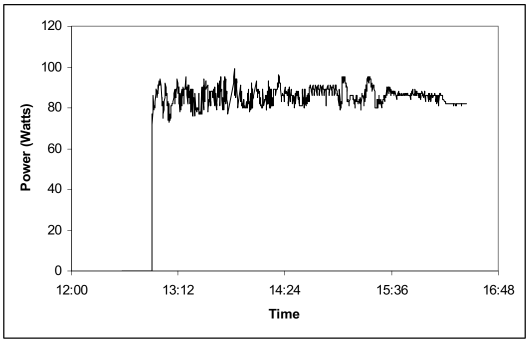

Fig.5 Ultrasonic power versus time in an extraction trial carried out at 250 bar, 60ºC and 15 kg/h

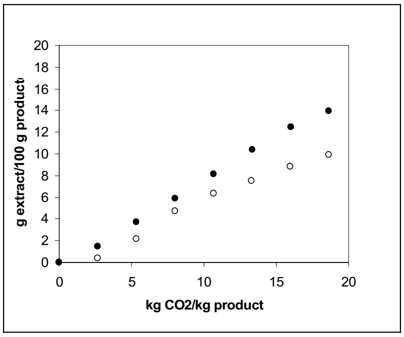

Fig.6 Almond-oil extraction curve at 320 bar and 45ºC with (●) and without (○) ultrasound

3.2. Almonds-oil extraction trials

In order to study the effects of USFE in almond-oil extraction, trials were carried out at various pressures (200 – 320 bar), two extraction temperatures (45 and 60ºC), times (up to 3.5 h) and CO2 flow rates (10 – 15 kg/h). The effect of the extraction pressure was study. At 200 bar the improvement in the yield extracted (mass transfer) was only 15% probably due to the low solubility of the almond-oil in the supercritical CO2. However, the yield extracted from grounded almond at 320 bar, 45ºC and 10 kg/h gave rise to 40% larger yields when HPU were applied as it is shown in Fig. 6 and 7. Even larger improvements between extraction curves with and without ultrasounds where achieved on experiments carried out at 280 bar, 45ºC and 12.5 kg/h extraction yields improvements of about 90% were obtained.

3.3. Cocoa cake-oil extraction trials

A third set of trials was carried out with a different substrate, cocoa beans. Solid samples were prepared for the experiments as follows: initial substrate was milled and sieved before the treatment in order to have a particle size distribution between 2 – 3.5mm. Next, the samples were placed in the basket inside the extractor to begin with the extraction trials at 320 bar and 65ºC. Good results were also obtained in the oil extraction with ultrasound. In fact, as shown in Fig.8, the application of the ultrasonic energy increases the extracted yield in around 43%.

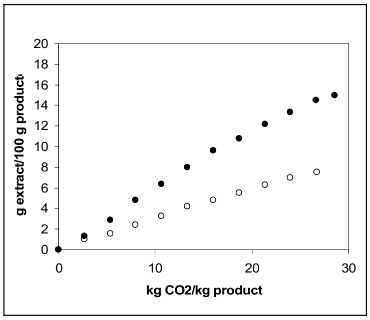

Fig.7 Almond-oil extraction curve at 280 bar and 45ºC with (●) and without (○) ultrasounds

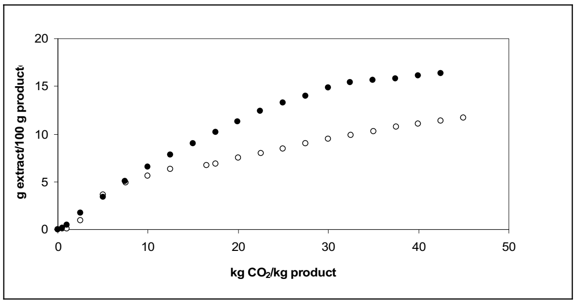

Fig.8 Cocoa cake-oil extraction curve at 320 bar and 65ºC with (●) and without (○) ultrasounds

4. Conclusions

A new system for ultrasonic application in supercritical extraction processes at pilot plant scale was designed, build-up, tested and validated. The power ultrasonic system operates in an automatic way during the extraction process. It means that it is not required any manual intervention by external operator and the process can be carried out with good performance, reliability and stability. This fact represents a relevant innovation with regard to previously developed ultrasonic device. The HPU also demonstrated that the enhancement in the extraction results with the application of the ultrasonic energy by using the new system is higher than previously reported results. In fact, by using the previous ultrasonic system improvements of about 20% in almond-oil extraction yields were achieved, while with the present system the improvements reached up to 90%., The USFE new system has also been used with other substrates as cocoa cake obtaining enhancements in the yield extraction of about 43%.

Acknowledgements

Work supported by the National Research Project PETRI-PTR95-075.OP.02. The authors would like to thank V.M. Acosta for his collaboration in the design of the power transducers by FEM and their development.

References

[1] C. Jun, Y. Kedie, C. Shulai, T. Adschiri and K. Arai, 4th International Symposium on Supercritical Fluids, 11-14 May, Sendai, Japan, (1997) 707.
[2] E. Riera, Y. Golás, A. Blanco, J.A. Gallego, M. Blasco and A. Mulet, Ultrasonics Sonochemistry 11 (2004) 241.
[3] S. Balachandran, S.E. Kentish, R. Mawson and M. Ashokkumar, Ultrasonics Sonochemistry 13 (2006) 471.
[4] A. Hu, S. Zhao, H. Liang, T. Qiu and G. Chen, Ultrasonics Sonochemistry 14 (2007) 219.
[5] M.D. Macías-Sánchez, C. Mantell, M. Rodríguez, E. Martínez de la Ossa, L.M. Lubian and O. Montero, Talanta 77 (2009) 948.
[6] T. Mason, E. Riera, A. Vercet and P.Lopez-Buesa, Chapter 13 "Applications of Ultrasound" in Emerging Technologies for Food Processing, ed Da-Wen Sun, Elsevier (2005) 323.
[7] M. Vinatoru, Ultrasonic Sonochemistry 8 (2001) 303.
[8] E. Riera, J.A. Gallego, F. Montoya, A. Blanco, A. Mulet, J.J. Benedito, R. Peña, Y. Golás, A. Berna, S. Subirats, M. Blasco and J. García, (2004) EP 1547679 A1.
[9] E. Riera, A. Blanco, V.M. Acosta, J.A. Gallego-Juárez, M. Blasco and A. Mulet, Proceedings of the 19th International Congress on Acoustics, ICA2007, Madrid (Spain), 2-7 September 2007.
[10] S.L. Taylor, N.B.A. Higley and R.K. Bush, Advances in Food Research 30 (1986) 2.
[11] E. Riera, J.A. Gallego, A. Blanco and V.M. Acosta, Proceedings of the International Congress on Ultrasonics, ICU2007, Paper 1435, Vienna (Austria), 9-13 April 2007.