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Optimizations and comparison of two supercritical extractions of adlay oil

Aijun Hu [*,1], Zhihua Zhang, Jie Zheng, Yiming Wang, Qiongxi Chen, Rong Liu, Xue Liu, Shujing Zhang

Key Laboratory of Food Nutrition and Safety (Tianjin University of Science & Technology), Ministry of Education, College of Food Engineering and Biotechnology, Tianjin University of Science & Technology, Tianjin 300457, P.R. China

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ABSTRACT

Oil is an important component of adlay seed (*Coix lachrymal-jobi L. var. Adlay*) with many beneficial functions to human health. In this work, a supercritical fluid extraction (SFE) and a novel ultrasonic-assisted supercritical fluid extraction (USFE) were studied and compared. Their operating conditions on the oil extraction, including extraction temperature (T), pressure (P), time (t), and CO_2 flow rate (F), were optimized. Based on the yield of extraction, the favorable conditions for SFE were: T at 45 °C, P at 25 MPa, t at 4.0 h and F at 3.5 L/h. While ultrasound was applied as in USFE, the following parameters were preferred: T at 40 °C, P at 20 MPa, t at 3.5 h and F at 3.0 L/h, respectively. The results show that supercritical fluid extraction with the assistance of ultrasound could reduce the temperature, pressure, CO_2 flow rate, as well as time used in the process. Compared with SFE, USFE could give a 14.0% increase in the yield for extracting oil from adlay seed with less severe operating conditions. According to the quality of adlay oil and the analysis of its fatty acids by GC-MS, there was no significant difference between USFE and SFE.

Industrial relevance: Adlay oil is an important component of adlay seed (*Coix lachrymal-jobi L. var. Adlay*) with many beneficial functions to human health. In China, there is already this kind of product produced by factories. However, its production method is conventional solvent extraction. Although supercritical fluid extraction has obvious advantages compared to conventional extraction method, it has also some disadvantages such as low extraction efficiency for some components, very high operation pressure and safety problem. In our work, a novel ultrasonic-assisted supercritical fluid extraction (USFE) was studied and compared. As an emerging technology, it has huge potential and remarkable significance in its application in adlay oil and other relevant products' production at factory-scale. The experimental results and conclusion will provide a useful background to application of the novel technology in industrials.

1. Introduction

Wild *coix lachrymal-jobi L.* (Coix) is native to and extensively grown in South Asia (Arora, 1977). The cultivated variety, *coix lachrymal-jobi L. var. adlay* (Adlay), is a soft-shelled seed crop cultivated in countries such as India (Arora, 1977), Brazil (Ottoboni, Leite, Targon, Crozier, & Arruda, 1990), Japan (Kondo, Nakajima, Nozoe, & Suzuki, 1998) and China (Kuo, Chiang, Liu, Chien, Chang, Lee, et. al., 2002). Adlay has long been used as an animal feed, as food for humans and in herbal medicine. It contains abundant active components such as adlay oil which has lots of important health and medical functions. Adlay oil can inhibit the growth of cancer cells with an efficiency of above 87%, and help prevent the decrease of white blood cells during chemical therapy (Lu, Zhang, & Zhang, 1999).

Normally adlay oil is obtained using mechanical or chemical processes. Mechanical processes often associate with low yields, while chemical extraction methods often involve the use of organic solvents which can be harmful to human health and environment (Mustakas, 1987). Tough new regulatory requirements on the use of organic solvents have prompted active research on clean extraction technologies (Reverchon & Osseo, 1994). Supercritical fluid extraction (SFE) is one of the newly emerging clean and environmentally friendly technologies for food and pharmaceutical products (De Azevedo, Kopcak, & Mohamed, 2003).

Among supercritical fluids, CO_2 is the most commonly used solvent for the extraction of oils from natural products. However, the efficiency of SFE is hindered by the low solubility of the triglycerides in CO_2, and the high pressure and long extraction time required (Reverchon & Osseo, 1994).

Ultrasound, a kind of elastic mechanical wave, can produce thermal effect, mechanical fluctuant effect and cavitation effect. Cavitation effect would cause the formation, the growth, the compression and the explosion of bubbles in the solution, which can lead to the dispersion of the solid particles, the increase of the contact area between the particles and extraction solvent, and the improvement of mass transfer rate from solid phase to liquid phase (Mantysale & Mantysalo, 2000; Velickovic,

* Corresponding author at: College of Food Engineering and Biotechnology, Tianjin University of Science & Technology, No. 29, 13th Avenue, Tianjin Economical-Technological Development Area, Tanggu district, Tianjin 300457, P. R. China. Tel.: +86 13512056805; fax: +86 22 60601445.
E-mail address: huaijun@tust.edu.cn (A. Hu).

[1] Research orientation: application of physical field techniques in food science and engineering, has already published nearly 100 papers, 6 books and applied 12 patents.

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Milenovic, & Ristic, 2006; Zheng & Hu, 2007). The ultrasound can effectively increase the extraction rate and speed up the extraction process.

Recently, the application of ultrasound techniques in solvent extraction has attracted more attention. The solvent extraction of oil from oil-tea camellia seed, soybean and flaxseed (Hu, Feng, & Zheng, 2009, 2002, Hu, Qiu, & Liu, 2002; Zhang & Zhao, 2006), as well as the extraction of functional ingredients from the Chinese herbal medicine(Ben & Qiu, 2006; Li, Pordesimo, & Weiss, 2004; Zeng & Qiu, 2005; Zhang, Wang, & Li, 2008), was found to be significantly improved by the introduction of ultrasound wave, and extensive researches on ultrasound assisted solvent extraction have been conducted.

At present, a great deal of researches on supercritical fluid techniques have been done, including SFE, and its applications in preparation of microparticles, nanoparticles recrystallization and dissolution (Chattopadhyay & Gupta, 2001, 2002; Enokida, Abd El-Fatah, & Wai, 2002; Han, Zhang, & Cheng, 2007; Jia, Lu, & Sun, 2007). However, the application of ultrasound in SFE is still in the developing era. Sethuraman studied ultrasound-assisted SFE (USFE) of capsaicin from peppers, the experimental results indicated the extraction amount and the loading amount of extraction vessel of USFE were higher than those of SFE (Sethuraman, 1997). Trofimov used ultrasound during the dissolution of uranium oxides in supercritical carbon dioxide. The results showed the dissolution of uranium trioxide with sonification was increased by approximately 100% at 60 °C and 150 atm (Trofimov, Samsonov, Lee, Smart, & Wai, 2001). More recently the use of power ultrasound in SFE to enhance the extraction yield of oil from almonds was reported by Riera, Golas, Blanco, Gallego, Blasco, & Mulet (2004), and he found that power ultrasound significantly accelerated the kinetics of the process and improved the final extraction yields by 20%, due to the effects produced by ultrasound, such as the compression, the decompression, radiation pressure and acoustic streaming. Till now, the optimal SFE and USFE conditions of adlay oil have not been reported, and the effects of ultrasound in SFE on the quality and components of adlay oil have also not been studied.

The main objective of this work is to extract adlay oil using SFE with and without ultrasound. The results obtained from USFE will be compared with that obtained from the SFE to reveal the effects of ultrasound under supercritical conditions.

2. Materials and methods

2.1. SFE and USFE system

The SFE equipment was designed by Guangzhou Light Engineering Institute. The USFE system was self-designed based on the SFE equipment. As showed in Fig. 1, the USFE system is made up of two units: SFE unit and ultrasonic unit. The major components of SFE include a positive displacement liquid pump, a 1000 mL pressurized extraction vessel, a separation column and a separation vessel. The extraction vessel and two separators are equipped with water jackets and temperature controllers. In order to ensure accurate and stable supercritical CO_2 delivery, the pump head is cooled by circulating water. The temperatures in the system are controlled within ± 0.1 °C and the pressures within ± 0.5 MPa.

The probe with Langevin type transducer is installed in the upper part of the extractor, and driven by electrical signals from an ultrasound generator, which gives adjustable continuous power outputs at fixed frequency of 20 kHz. The transducer can stand for a maximum temperature of 105 °C. The ultrasound generator consists of a power amplifier and a special electro circuit designed to justify the power outputs at a constant level during the USFE process. The electroacoustical efficiency of the ultrasonic transducer is 87%.

2.2. Materials

Adlay seeds were bought from the local traditional Chinese medicine shop. The oil content of the adlay seed, as determined by extraction with acetone, was found to be 9.5%. CO_2 of 99.9% purity was obtained from Guangzhou Gas Company, China. All chemicals were purchased from local chemical stores and at analytical grades.

2.3. Experimental methods

In order to fully compare the effects of USFE and SFE, experiments were carried out at various extraction temperatures, pressures, times and CO_2 flow rates. 100 g of grounded adlay seeds was placed into the extractor in a typical extraction experiment. Liquid CO_2 was pumped into the extractor until the desired extraction pressure was reached. The extractor was heated to the extraction temperature, and pressure valves located downstream of the extractor were slowly opened while maintaining the pressure constant in the extractor. When the set conditions reached, the extraction process started to time. The adlay oil, which precipitated under a low pressure and temperature in the separation vessel, was recovered, when extraction time reached.

2.4. Calculation of adlay oil extraction yield

The adlay oil extraction yield (EY) is calculated using the following formula:

$$EY = m_t / m_0 \times x_0 \times 100 \quad (1)$$

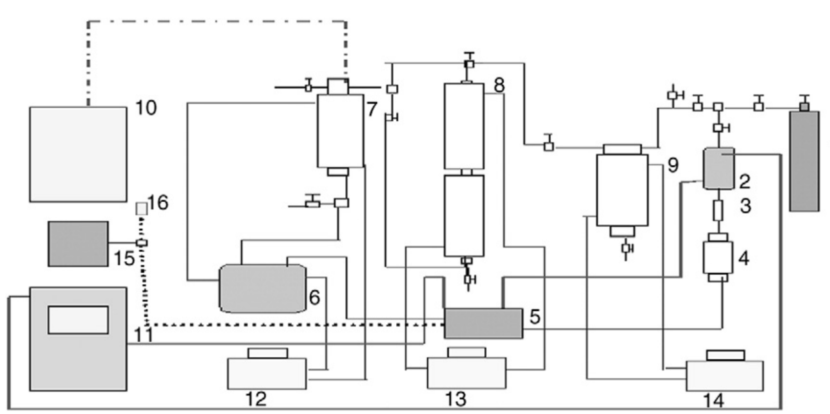

Fig. 1. Ultrasonic-assisted supercritical fluid extraction apparatus[2]

[2] 1. CO_2 steel cylinder (vessel); 2. Cold trap; 3. Flowmeter; 4. CO_2 deposition tank; 5. CO_2 pressure pump; 6. Heat exchanger; 7. Extraction vessel; 8. Separation column; 9. Separation vessel; 10. Ultrasound generator; 11. Cooled engine; 12, 13, 14. Thermostatic water-bath heater; 15. Modifier pump; 16. Modifier vessel.