Journal of Food Engineering 105 (2011) 609–616

Contents lists available at ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

A supercritical tuneable process for the selective extraction of fats and essential oil from coriander seeds

Houcine Mhemdi [a,b], Elisabeth Rodier [a], Nabil Kechaou [b], Jacques Fages [a,*]

[a] Université de Toulouse, Ecole des Mines Albi, RAPSODEE Centre, CNRS, F-81013 Albi, France
[b] Université de Sfax, Ecole Nationale d'Ingénieurs de Sfax, Groupe de Recherche en Génie des Procédés Agroalimentaire, Unit MFAM, 02/UR/11-02, 3038 Sfax, Tunisia

ARTICLE INFO

Article history:
Received 3 August 2010
Received in revised form 25 March 2011
Accepted 29 March 2011
Available online 3 April 2011

Keywords:
Coriander seeds
Vegetal oil
Essential oil
Supercritical CO_2
Selective extraction

ABSTRACT

A selective supercritical CO_2 extraction process has been developed in order to extract selectively the vegetal oil and the essential oil from coriander seeds in a consecutive way by tuning experimental conditions. A 4-step process has been set up: (i) seeds preparation by grinding and sieving, (ii) extraction by supercritical CO_2 and (iii; iv) selective separation in two separators with different pressure levels. The first part of this study consisted in studying the effects of the operating conditions (T, P, CO_2 density, flow rate and particle size distribution of ground seeds) on the yield of extraction. The results showed that the global performance of the process depends mainly on the pressure level of extraction, on the CO_2 flow rate and on the size of the ground seeds. An optimization of the parameters led to an extraction ratio of 90%. The second part of this study dealt with the effects of the operating conditions on the quality of extracts, and more precisely on their composition in vegetal oil and essential oil. It was observed that the supercritical extraction had no major effect on the fatty acid composition of the vegetal oil compared to classical extraction techniques, while essential oil is enriched in components such as linalool and that a previous drying of the seeds is deleterious. Finally, we propose a procedure in order to extract separately each oil by adjusting CO_2 density and separation conditions.

1. Introduction

Coriander (*Coriandrum sativum* L.) is an annual *Apiaceae* (*Umbelliferae*) herb, which is widely used in food (Aluko et al., 2001; Burdok and Carabin, 2009), pharmaceutical (Jabeen et al., 2009) and cosmetic (Eyres et al., 2005) industries.

Coriander fruits contain vegetal oil (VO) with a high concentration of monounsaturated fatty acids, especially of petroselinic acid (C18:1 cis-Δ^6). This acid can be oxidatively cleaved to produce a mixture of lauric acid (C12:0), a compound useful in the production of detergents, and adipic acid, a C6 dicarboxylic acid, which can be utilized in the synthesis of nylon polymer (Msaada et al., 2009b). Moreover, the essential oil (EO) of coriander has been shown to have antibacterial (Kubo et al., 2004; Matasyoh et al., 2009), antioxidant (Wangensteen et al., 2004), antidiabetic (Gallagher et al., 2003), anticancerous and antimutagenic (Chithra and Leelamma, 2000) activities.

There are three major extraction techniques used to obtain VO and EO from coriander seeds, which are steam distillation, organic solvent extraction (Soxhlet), and supercritical fluid extraction. Each technique has its benefits and drawbacks as far as operating cost, capital cost, yield and quality of the extracts are concerned. Steam distillation is by far the most widely used and the cheapest way of extraction, but it is limited to EO production, and it may induce chemical changes in the extract by oxidation of some compounds (Anitescu et al., 1997; Donelian et al., 2009; Msaada et al., 2007). Organic solvent extraction is intermediate in capital and operating cost, and is used for producing VO from oilseeds. However, concerns about the solvent residues in the oleoresin products, the new regulations of volatile organic solvent emissions in the air, and the extent of further refining that is required after the extraction step restrain the use of this technology (Catchpole and Grey, 1996). Supercritical fluid extraction is the most recent technology that is of increasing importance in the production of EO, VO, and a range of other substances from natural products (Brunner, 2005; Mohamed and Mansoori, 2002; Temelli, 2009). This technique has the highest capital cost of the three techniques, and moderate operating costs. However, neither solvent residues remain in the product after extraction, nor there are any chemical

Abbreviations: Bi, Biot number; *d*, diameter (m); *D*, diffusivity (m^2/s); EO, essential Oil; ER, extraction ratio (%); EY, extraction yield (%); FA, fatty acid; *K*, mass transfer coefficient (m/s); *m*, mass of the extract (g); MUFA, monounsaturated fatty acid; *P*, pressure (Pa); PUFA, polyunsaturated fatty acid; *Q*, flow rate of CO_2 (kg/h); FA, saturated fatty acid; *T*, temperature (°C); VO, vegetal oil; ρ, CO_2 density (kg/m^3); σ, standard deviation.

* Corresponding author. Tel.: +33 (0)5 63 493 141; fax: +33 (0)5 63 493 025.
E-mail address: jacques.fages@mines-albi.fr (J. Fages).

0260-8774/$ - see front matter
doi:10.1016/j.jfoodeng.2011.03.030

changes due to the processing technique, which gives extract of outstanding quality (Boutin and Badens, 2009; Brunner, 2005; Catchpole and Grey, 1996; Machmudah et al., 2008; Donelian et al., 2009; Perrut and Clavier, 2003).

The review of literature shows that most of the research works related to the extraction of coriander seeds are aimed at extracting either the VO (Msaada et al., 2009a,b) or the EO (Bandoni et al., 1998; Grosso et al., 2008; Illés et al., 2000; Msaada et al., 2007). The aim of this work is to put forward a new process using supercritical carbon dioxide, which is able to extract selectively the VO and the EO in a consecutive way by tuning experimental conditions. The effects of operating conditions on the yield of extraction and on the quality of extracts will be also studied to optimize the process.

2. Materials and methods

2.1. Raw material

Coriander seeds came from Canada and were provided by the laboratory of General Herbalism, Marseille, France. The seeds used were mature and brown. They were stored in a refrigerator at 4 °C before use. The moisture content was about 10%. Their mass content of VO and EO, respectively determined by Soxhlet and steam distillation were 20.8% (w/w) of VO and 0.4% (w/w) of EO. Hence, EO represents 1.9% (w/w) of the total oil content. The crushed coriander seeds (mean size 0.3 mm) were used for Soxhlet and steam distillation extraction of EO and VO. The extraction was carried for a long time (6 h for VO extraction and 90 min for EO extraction) to ensure total oil extraction. The supercritical extraction fluid was carbon dioxide (purity 99.995%, w/w) purchased from Air liquide S.A., Paris, France.

2.2. Particle size reduction

In mature coriander seeds, the oil is homogeneously distributed and it is bounded by cell membranes. Crushing the seeds may damage some cell membranes leading to the release of some oil and facilitating the extraction step. To study the effect of particle size on the extraction ratio, coriander seeds were first crushed. The grinding was performed using a grinder type IKA M20. The particle size distribution of ground seeds was determined by sieving. Size fractions were then obtained using sieves with an aperture size of 200 μm, 400 μm, 630 μm, 800 μm and 1 mm. Afterwards, three different fractions were used for extraction, having a mean size of 0.3 mm, 0.515 mm (named 0.5 in the following sections) and 0.9 mm.

2.3. Experimental set-up and protocol for extraction

Supercritical CO_2 extraction experiments on coriander seeds were carried out on the pilot scale apparatus (Separex, France) shown schematically in Fig. 1. The mass of ground and sieved coriander seeds used in each experiment was approximately 50 g.

The pilot scale tests were performed by flowing carbon dioxide at the required temperature and pressure through a bed of ground coriander contained in the extractor (8). A backpressure regulator controlled the pressure in the extractor and temperature was regulated by an electrical device. The carbon dioxide and dissolved solute was then depressurized causing desaturation and condensation of VO into the first cyclonic separator (S1), which was held at a pressure of 7 MPa and a temperature of 30 °C. Then carbon dioxide flowed through a needle valve causing desaturation and condensation of the remaining EO and water into the second cyclonic separator (S2). This last vessel was maintained at a pressure of 4 MPa and a temperature of 10 °C. After the extraction step, a yellow VO, was recovered in separator 1 (S1) and a fragrant EO is collected in separator 2 (S2). Extracts were sampled every 15 min from the separation vessels in order to follow the kinetics of extraction. The experiment was stopped when the amount extracted remained constant (i.e. once the derivative of the mass of extract in function of time, dm/dt, was lower than 0.005 g/min) and a plateau was reached on the extraction kinetics.

2.4. Chromatographic analysis

2.4.1. Fatty acid composition of vegetal oil

The fatty acid composition of the coriander VO was determined by analysis of their methyl esters obtained by transesterification. These esters were analyzed using a gas chromatograph type Varian CP-select CB 3900 equipped with a silica capillary column (50 m × 0.25 mm × 0.25 μm). Temperature was held at 185 °C for 4 min, raised to 250 °C at 15 °C/min. The injection and the detector temperatures were both 250 °C. The carrier gas was H_2 (1.2 mL/min). The device was equipped with a FID detector (lowest detectable quantity: 5 pg of C), which gave the chromatogram for each sample. The identification of fatty acids was made by comparing their retention times with those of standard samples. Their mass content was calculated by assuming that the total peaks area account for 100% of the constituents.

2.4.2. Composition of essential oil

The separation of volatile components of the coriander EO was performed using a gas chromatograph capillary type HP 5890 series II, equipped with a DB-5MS column (30 m × 0.25 mm × 0.25 μm), a detector of flame ionization (FID) and injector EPC (electronic pressure control). The tests were conducted according to the following conditions. The column was held at 40 °C for 5 min and then increased at 5 °C/min up to 280 °C over 53 min. The solvent was dichloromethane and the flow through the column was 1.3 mL/min. The FID detector was held at 300 °C. The head pressure of the column was 0.1 MPa and the amount injected was 0.5 μL. The identification of the different components of EO was made by calculating the retention index and Kovats index and by comparing them with those of the literature.

2.5. Theory and calculation

2.5.1. Extraction yield and extraction ratio

In this study, the extraction yield (EY) is defined as the total mass of extract recovered from 100 g of coriander. It is expressed in% (g of extract/100 g coriander). The extraction ratio (ER) is the ratio in mass percent of oil extracted over the total quantity of oil contained in the seeds of coriander.

2.5.2. Mass transfer parameters

The supercritical fluid extraction implies the mass transfer of the VO and EO from coriander seeds to the flowing carbon dioxide. It is then important to understand this phenomenon and to appreciate its extent. This requires to estimate the parameters associated to mass transfer, such as the diffusivity of the components to be extracted, D, resulting from the diffusive internal transfer inside the seeds, the mass transfer coefficient, K, resulting from the convective transfer at the seeds surface and the Biot number, Bi, comparing both transfers.

The model used in this work to describe supercritical carbon dioxide extraction of oils from coriander seeds was developed by Goto et al. (1993, 1994). It is applicable to both essential and vegetal oil. This model considers coriander seeds like a porous solid containing EO associated with lipids located in vesicles (subsequent pores when opened). Internal mass transfer of oils implies