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Supercritical fluid extraction of black pepper oil

C. Perakis *, V. Louli, K. Magoulas

Laboratory of Thermodynamics and Transport Phenomena, School of Chemical Engineering, National Technical University of Athens, 9, Heroon Polytechniou Str., Zografou Campus, 15780 Athens, Greece

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Abstract

The supercritical fluid extraction (SFE) of oil from ground black pepper, using supercritical carbon dioxide (SC CO_2) as a solvent, is presented in this study. The effect of process parameters, namely pressure (90, 100, 150 bar) and temperature (40, 50 °C) of extraction, and solvent flow rate (1.1, 2, 3 kg/h), on the extraction rate was examined in a series of experiments conducted in a bench scale apparatus. The results indicated a significant increase of extraction rate with increase of pressure or decrease of temperature. A similar effect was observed with the increase of solvent flow rate. The experimental data were satisfactorily correlated by two mass balance models. The first one is based on the Lack's plug flow model, which accounts for both the solubility and diffusion controlled regimes of the extraction, and the second one on the adsorption–desorption equilibrium of solute from solid tissue, the diffusion of the solute dissolved in the supercritical solvent to the surface and the mass transfer through the external film into the bulk.

Keywords: Supercritical fluid; Extraction; Black pepper oil; Process parameters; Mass transfer; Modelling

1. Introduction

Pepper, a well-known pungently aromatic condiment, is the dried berry, either whole or powdered, of the *Piper nigrum*, a vine that can grow up to 10 ft. tall and is indigenous to India and Asia. In general, common or black pepper is made from the whole berry, dried just before maturity and it is an important spice, appreciated for both its aroma and its pungency. Volatile components of pepper's oil contribute to its aroma, while alkaloid compounds to its pungency (Bauer, Garbe, & Surburg, 1997; Govindarajan, 1977).

Pepper oil can be stored more easily and safely than ground pepper (Govindarajan, 1977). Therefore there is much interest in its extraction procedure. Lately, SFE has gained increasing attention over the traditional techniques, like steam distillation and liquid solvent extraction, as the use of a non-toxic and volatile solvent, such as CO_2, protects extracts from thermal degradation and solvent contamination (Brunner, 1994; McHugh & Krukonis, 1986). Thus, various studies on SFE to obtain black pepper oil have been reported (Ferreira, Nikolov, Doraiswamy, Meireles, & Petenate, 1999; Sankar, 1989; Sovová, Jež, Bártlová, & St'astová, 1995; Tipsrisukond, Fernando, & Clarke, 1998; Vidal & Richard, 1987).

Despite the number of SFE experimental data reported for many natural species including pepper, the modelling and simulation of the SFE process remains a major issue of research activity, justifying the need of more reliable experimental data.

So far, the extraction process has been described by empirical models (Naik, Lentz, & Maheshwari, 1989; Nguyen, Barton, & Spencer, 1991), models based on heat transfer analogy (Bartle et al., 1990; Reverchon, Donsi, & Sesti Ossèo, 1993), by assuming that the vegetable particle is a single sphere, which is cooled in

* Corresponding author. Tel.: +30 210 772 3230; fax: +30 210 772 3155/+30 353 2 716 7714.
E-mail address: cperakis@central.ntua.gr (C. Perakis).

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a uniform fluid medium, and finally mass balance models. The latter are the most flexible, and various authors have employed them, proposing different assumptions about mass transfer or equilibrium mechanisms controlling the SFE depending on the natural matrices examined. Thus, Bulley, Fattori, Meisen, and Moyls (1984) assumed that the mass transfer resistance in the solvent phase controlled the extraction, while Reverchon (1996) found that the internal diffusion was the dominant phenomenon in SFE of sage. King and Catchpole (1993) employed a shrinking core model. Sovová (1994) and Sovová, Kučera, and Jež (1994) introduced the theory of broken and intact cells of the solid phase, and took into account both external and internal mass transfer resistance. Also, Goto, Sato, and Hirose (1993) and Peker, Srinivasan, Smith, and McCoy (1992) considered that SFE involved an adsorption–desorption mechanism. Finally, many authors employed further assumptions concerning the shape and structure of the particles (Goto et al., 1993; Reverchon, 1996; Reverchon & Marrone, 2001; Roy, Goto, & Hirose, 1996), the equilibrium expression (Goto, Roy, Kodama, & Hirose, 1998; Perrut, Clavier, Poletto, & Reverchon, 1997), etc.

Thus, it becomes obvious that a single model cannot describe all the experimental data available, due to the different behaviors of different vegetable matrices. Consequently, more research is needed in order to obtain a better understanding of the SFE process of natural species.

In this work, the influence of pressure, temperature and solvent flow rate on the extraction rate of ground black pepper was studied. The process was also described by two mass balance models: (i) the extended Lack's plug flow model proposed by Sovová (1994) and Sovová et al. (1994), and (ii) a model based on the adsorption–desorption mechanism coupled with the linear driving force (LDF) concept (Škerget & Knez, 2001).

2. Materials and methods

2.1. Materials

Black pepper berries, supplied by VIORYL S.A. (Greece), were ground and sieved prior to the extraction. The mean particle diameter (d_p) was 175 μm. The CO_2 (99.5 wt%) was supplied in liquid state by Air-Liquide (Greece) and the reference standard piperine by Acros Organics.

2.2. Experimental apparatus and procedure

The experiments were carried out in a bench scale apparatus SFE-500 (SEPAREX, France) designed to conduct batch operations. A flow sheet of the apparatus, which is composed of a high pressure vessel (400 ml) and two high performance cyclonic separators (18 ml each) operating up to 300 bar, is presented in Fig. 1. A more detailed description of the apparatus is given elsewhere (Papamichail, Louli, & Magoulas, 2000).

In each experiment a quantity of ground berries equal to 100 g was placed between two layers of glass beads (d = 4.5 mm), which were added to reduce the dead space in the extractor vessel and allow the uniform distribution of the solvent flow. Extraction yield and rate were obtained by measuring the weight loss of the feed at regular periods of time. The operating conditions of each experiment are reported in Table 1.

Pressure effect was studied at 40 °C and 2 kg CO_2/h, while the examined extraction pressures were 90, 100 and 150 bar. Temperature effect was examined at

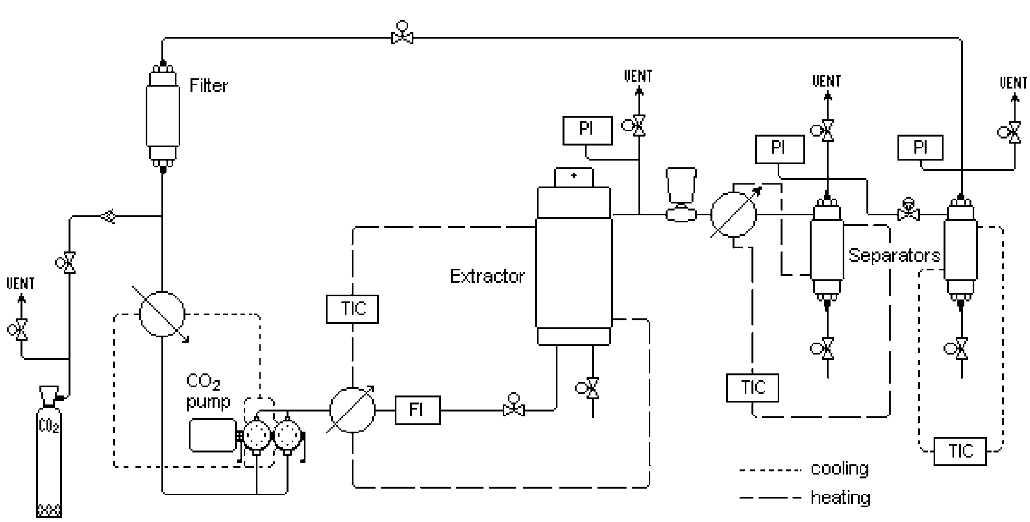

Fig. 1. Flow sheet of the bench scale apparatus (SFE-500).