

Research note

Vetiver Oil Extraction Optimization Using Supercritical Carbon Dioxide Fluid

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Abstract

This paper reports a study to find optimum conditions for oil extraction from vetiver root. For this purpose, the influence of temperature and pressure on the extraction yield is investigated. In addition, the effects of supercritical fluid flow rate and particle diameter on optimum temperature and pressure have been studied. The results show that optimum pressure is a strong function of particle diameter and solvent flow rate. However, the results reveal that the optimum temperature is independent from particle diameter and solvent flow rate.

Keywords: *Optimization; Supercritical Fluid Extraction; Vetiver Oil*

1- Introduction

Increasing environmental concerns regarding the use of organic solvents in the extraction of natural products have encouraged the growing interest in supercritical fluid applications [1-5]. The supercritical fluid extraction (SFE) eliminates the disadvantages of conventional solvent extraction which leads to degradation of heat sensitive compounds and leaves traces of toxic solvents in the solute.

There are several experimental data for SFE from solid substratum, in which the material extract using SFE was compared with extracts obtained from other techniques [6]. One of the studied plants is the root of vetiver due to its commercial importance.

Vetiver (*Vetiveria zizanioides* (L.) Nash ex Small) is found in tropical regions, such as India, China, Indonesia, Haiti and the Reunion Island, who are the world's main vetiver oil producers [7].

Martínez et al. [8] compared different methods of extracting oil from Brazilian vetiver roots. Their comparison was based on the extraction yields and the quality of the products. Meireles et al. [9] used application of a theoretical model for describing SFE extraction from vetiver roots and studied the influence of some process parameters such as supercritical fluid flow rate, extractor geometry, particle diameter and bed void fraction on the extraction yield.

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The main objective of the current study is to develop a theoretical model for extraction of vetiver oil and identify the optimum operational condition (T, P) by a new thermodynamic method. In addition, the influence of some parameters such as solvent flow rate and particle diameter on the optimum temperature and pressure has been studied. For verification of the model our previous experimental data have been used [9].

2- Mathematical modeling

2.1- Model and parameters

The proposed model of Meireles et al. [4] has been used for optimization. The density of CO₂ was calculated by Peng-Robinson equation of state [10]. The binary diffusion coefficient of solute in supercritical solvent, D_m, was obtained using Chao-Hong He correlation [11]. Other parameters such as viscosity, axial dispersion coefficient and Sherwood number were calculated using the same method that Meireles et al. [9] used. Furthermore, the calculation of Equilibrium constant (K) is explained in the following section.

2.2- Equilibrium constant calculation

In order to calculate the vetiver oil equilibrium constant between vetiver seed and supercritical carbon dioxide, the fugacity concept has been used. In equilibrium condition, the vetiver oil fugacity must be the same in each phase [12, 13]:

$$F_i^g = F_i^3 \quad (1)$$

Where i denotes the vetiver oil. The fugacity of solute in the fluid phase is [12, 13]:

$$F_i^g = y_i P \exp \left(\frac{1}{RT} \int_0^P \left[\left(\frac{\partial V}{\partial n_i} \right)_{T,P,n_2} - \frac{RT}{P} \right] dP \right) = y_i P \Phi_i \quad (2)$$

The dimensionless Φ_i is called the fugacity coefficient, which is calculated using an equation of state (such as Peng-Robinson). Fugacity of a component in solid phase at given temperature, T, pressure, P, and mole fraction, x_i has been calculated as follows [12]:

$$F_i^s = x_i P \exp \left(\frac{1}{RT} \int_0^P \left[\left(\frac{\partial V}{\partial n_i} \right)_{T,P,n_2} - \frac{RT}{P} \right] dP \right) \quad (3)$$

By separating the integral of equation (3) into two parts, the following equation can be obtained:

$$F_i^s = x_i P \exp \left(\frac{1}{RT} \int_0^{\alpha P_i^{sat}} \left[\left(\frac{\partial V}{\partial n_i} \right)_{T,P_i,n_{j,i}} - \frac{RT}{P} \right] dP \right) + \frac{1}{RT} \int_{\alpha P_i^{sat}}^P \left[\left(\frac{\partial V}{\partial n_i} \right)_{T,P_i,n_{j,i}} - \frac{RT}{P} \right] dP \quad (4)$$

The first and second integrals calculate the solute fugacity at vapor and condensed phases, respectively. In addition, α is a correction factor which is used to show the influence of insoluble solid phase on the solute vapor pressure. At low pressures, ideal gas equation can be used, so:

$$\left(\frac{\partial V}{\partial n_i} \right)_{T,P_i,n_{j,i}} = \frac{RT}{P} \quad (5)$$

At high pressure, the vetiver oil mixed with the insoluble solid phase completely, so the vetiver oil mole change has no effect on the solid volume, then:

$$\left(\frac{\partial V}{\partial n_i}\right)_{T,P,n_{j \neq i}} = 0 \quad (6)$$

Combining equations (4), (5) and (6) gives:

$$F_i^g = x_i P \exp\left(\frac{1}{RT} \int_0^{\alpha P_i^{\text{sat}}} \left[\frac{RT}{P} - \frac{RT}{P}\right] dP + \frac{1}{RT} \int_{\alpha P_i^{\text{sat}}}^P \left[0 - \frac{RT}{P}\right] dP\right) \quad (7)$$

By simplifying the equation (7), the following equation can be obtained:

$$F_i^g = x_i \alpha P_i^{\text{sat}} \quad (8)$$

Combining equations (2) and (8) gives:

$$y_i \Phi_i P = x_i \alpha P_i^{\text{sat}} \quad (9)$$

Solute mole fractions in supercritical and solid phase are calculated as follows:

$$y_i = \frac{C_i}{C_g} = \frac{C_i}{\frac{P}{zRT}} = \frac{zRT}{P} C_i \quad (10)$$

$$x_i = \frac{q_i}{q_i + q_3} = \frac{q_i}{q_i + \frac{\rho_3(1-X_0)}{M_3}} \quad (11)$$

Combining equations (9), (10) and (11) gives:

$$\frac{zRT}{P} C_i \Phi_i P = \frac{q_i}{q_i + \frac{\rho_3(1-X_0)}{M_3}} \partial P_i^{\text{sat}} \quad (12)$$

This equation, on rearrangement, becomes:

$$q_i = \frac{zRT \Phi_i \rho_3 (1-X_0)}{M_3 \alpha P_i^{\text{sat}} - zRT \Phi_i M_3 C_i} C_i \quad (13)$$

At low solute concentration, equation (13) converts to the following equation:

$$q_i = \frac{zRT \Phi_i \rho_3 (1-X_0)}{\alpha M_3 P_i^{\text{sat}}} \Big|_{y_i=0} C_i \quad (14)$$

In order to use equation (13), two parameters, including the correction factor (α) and insoluble solid molecular weight (M_s), must be determined so that the model results satisfy the experimental data. However, only one parameter (αM_s) should be determined if equation (14) is to be used. Therefore, due to lack of data, it is better to use equation (14). On the other hand, using a linear correlation between q_i and C_i instead of a nonlinear one is a more efficient way for calculating the equilibrium constant, so the equilibrium constant is calculated as follows:

$$K = \frac{zRT \Phi_i \rho_3 (1-X_0)}{\beta P_i^{\text{sat}}} \Big|_{y_i=0} \quad (15)$$

Where:

$$\beta = \alpha M_3 \quad (16)$$

The “ β ” parameter should be determined to minimize the error between the model result and the constant residence time data. The algorithm for calculating “ β ” is similar to that of the Meireles et al. [9] algorithm which

has been used to calculate the equilibrium constant. The calculated value of “ β ” using this method is equal to 1204.4.

2.3- Genetic algorithm

Implementation of Genetic algorithm (GA) makes use of the Darwinian survival of the fittest procedure. GA are search procedures based on the mechanics of natural genetic and natural selection. In this paper, GA is applied to determine the optimal extractor operating conditions (temperature and pressure) which maximize the extraction yield. The procedure is:

1. Definition of the fitness function. In this paper the fitness function is the extraction yield.
2. Characterization of GA parameters (population size: 500, Generations: 100, Selection Options: Stochastic uniform, Crossover Options: Scattered, Crossover Fraction: 0.8 and Mutation Options: Gaussian).
3. Production of initial generation in a random way.
4. Fitness evaluation.
5. Reproduction of a new generation using the GA selection operator.
6. Obtaining crossover pairs of members in the new generation.
7. Performing mutation in the new generation.
8. Steps 4–7 should be repeated until the number of generation reaches the prescribed value [14-16].

3- Results and discussion

Finding a an optimum operational condition is a complicated task. The particle diameter, solvent flow rate, temperature and pressure

are the main operational conditions, which should be determined in order to maximize the amount of extract. The optimal particle size should be as small as possible, but not in an order that leads to small particles sticking and causes solvent channeling in the extractor. The optimal solvent flow rate should be as large as possible. In addition, the channeling problem must be considered here. Finally, finding an optimum temperature and pressure makes the problem more complicated [17].

Fig. 1 illustrates the influence of temperature and pressure on extraction yield (F). The optimum operational conditions are in conditions in which the extraction yield is maximized.

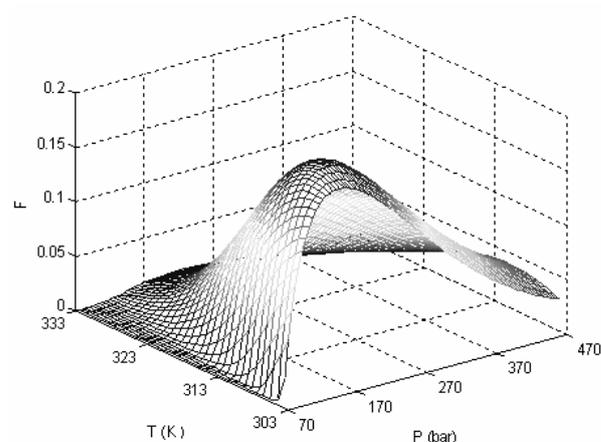


Figure 1. Influence of temperature (T) and pressure (P) on the extraction yield (F) with $\varepsilon = 0.699$, $Q = 10.5 \times 10^{-5} \frac{\text{kg}}{\text{s}}$, $L = 0.08\text{m}$, $D_{\text{ext}} = 0.0545\text{m}$, $d_p = 1.8 \times 10^{-4}\text{m}$ and $t = 10\text{min}$.

Using GA, the optimal condition, which resulted in the highest yield of vetiver oil, is obtained at a pressure of 167 bar and temperature of 307.75 K. Figures 2 and 3 reveal the influence of particle diameter and

solvent flow rate on the optimum pressure and temperature, respectively. Fig. 2 reveals that the optimum pressure decreases with an increase in particle diameter and increases with an increase in the solvent flow rate. On the other hand, as it is shown in Fig. 3, the optimum temperature is independent from the particle diameter and solvent flow rate.

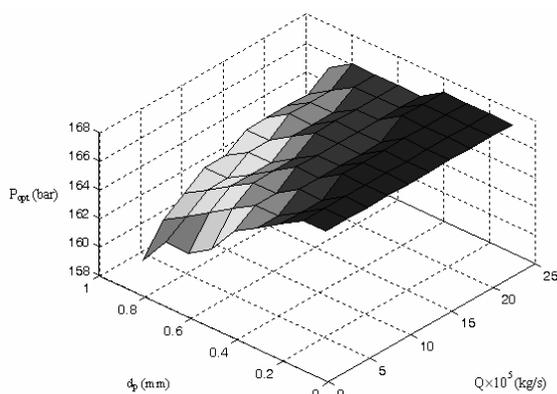


Figure 2. Influence of particle diameter (d_p) and solvent flow rate (Q) on the optimum pressure (P_{opt}) at $\varepsilon = 0.699$, $L = 0.08\text{m}$, $D_{ext} = 0.0545\text{m}$ and $t = 10\text{min}$.

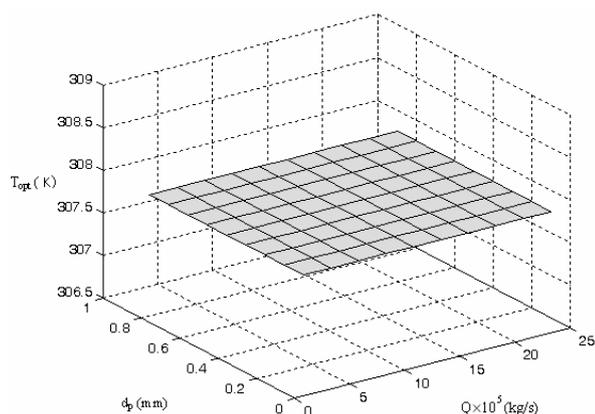


Figure 3. Influence of particle diameter (d_p) and solvent flow rate (Q) on the optimum temperature (T_{opt}) at $\varepsilon = 0.699$, $L = 0.08\text{m}$, $D_{ext} = 0.0545\text{m}$ and $t = 10\text{min}$.

4- Conclusions

In the present work, optimization of vetiver oil extraction using supercritical carbon dioxide has been studied. For this purpose, the vetiver oil equilibrium constant between the solid and supercritical phase was determined using the criterion of the equality of fugacity for each compound in both phases at equilibrium state. It has been found that the optimal condition which resulted in the highest yield of vetiver oil, at a fluid flow rate of $10.5 \times 10^{-5} \text{kg.s}^{-1}$, is a pressure of 167 bar and a temperature of 307.75 K. This optimum condition has been obtained using GA by 500 population size and 100 generations. In addition, the influence of supercritical fluid flow rate and particle diameter on the optimal conditions has been studied. The results indicated that optimum pressure decreases with an increase in particle diameter and increases with increasing the solvent flow rate. However, it has been found that the optimum temperature is independent on the particle diameter and solvent flow rate.

5- Nomenclature

$C, q \left(\frac{\text{kmol}}{\text{m}^2} \right)$	oil concentration in the supercritical and solid phase respectively
$d_p(\text{m})$	Particle diameter
$D_m \left(\frac{\text{m}^2}{\text{s}} \right)$	Molecular diffusion coefficient
F	Extraction yield, fugacity
K	Solute equilibrium constant between solid and fluid phase
L(m)	Extractor length
$m \left(\frac{\text{kg}}{\text{kmol}} \right)$	Molecular weight

$m_{\text{feed}}(\text{kg})$	Feed mass
$n(\text{kmol})$	mole
$P(\text{bar})$	Pressure
$Q\left(\frac{\text{kg}}{\text{s}}\right)$	supercritical fluid Flow rate
$R\left(\frac{\text{J}}{\text{kmol K}}\right)$	Gas constant
$T(\text{K})$	Temperature
$t(\text{min})$	Time
$V(\text{m}^3)$	volume
X	Solute mole fraction in solid phase
X_0	Initial mass fraction of solute in solid phase
y	Solute mole fraction in solvent
z	Compressibility factor

Greek letters

$\mu\left(\frac{\text{kg}}{\text{m}\cdot\text{s}}\right)$	viscosity
$\rho\left(\frac{\text{kg}}{\text{m}^3}\right)$	density
ε	Extractor void fraction
Φ	Fugacity coefficient
α	Correction factor

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