

Contents lists available at ScienceDirect

The Journal of Supercritical Fluids

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

Mathematical modelling of turmeric compounds extraction using high pressurized solvents mixture



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G R A P H I C A L A B S T R A C T



ARTICLE INFO

Keywords: Solvent mixture extraction Curcuma longa Kinetic Mathematical modelling

ABSTRACT

Mathematical modelling was carried out in order to study the kinetics of the sub- and supercritical fluid extraction from turmeric, using carbon dioxide with either ethanol or isopropyl alcohol and their mixture (1:1 v/v) as co-solvents. The variables studied were pressure, solvent flow rate, co-solvent percentage and bed height to bed diameter (H_B/D_B) ratio. Using the previously obtained experimental data, four mathematical models were evaluated in the present work to identify those that accurately fit the overall extraction curves. The models of Goto and Sovová were the ones that best correlated most of the experimental overall extraction curves obtained for turmeric extraction, even when using pressurized liquid solvent mixtures.

Sovová model k_{xa} Mass transfer in the solid phase $[1/s]k_{ya}$ Mass transfer coefficient in the fluid phase [1/s]tThe extraction time $[s]t_{CER}$ and t_{FER} Time related to the steps of CER and FER periods $[s]m_{IS}$ The initial mass of inert solid material $[kg]X_0$ The maximum extract obtained in the solid $(kg/kg)Y^*$ Extract solubility in the CO₂ $[kg/kg]x_k$ The mass fraction of easily accessible solute in the inert material $[kg/kg]\rho$ Density of CO₂ $(kg/m^3)\rho_p$ Density of the particles $(kg/m^3)\varepsilon$ Particles porosity [-]

1. Introduction

In the last years, several applications of supercritical fluid extraction (SFE) from turmeric (*Curcuma longa*) mainly using CO₂ as solvent, have been reported in the literature. The influence of pressure, temperature, and flow rate on the extract global yield was studied by Began et al. [1]. Gopalan et al. [2] measured the extraction rate as a function of pressure, temperature, flow rate, and particle size. The mass transfer of the

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https://doi.org/10.1016/j.supflu.2018.07.014

Received 29 March 2018; Received in revised form 10 July 2018; Accepted 11 July 2018 0896-8446/ © 2018 Elsevier B.V. All rights reserved.

Nomeno	Nomenclature		Effective diffusion	
		t	The extraction tim	
SFE	Supercritical fluid extraction	r	The particle radiu	
HB	Bed height [m]			
H_B/D_B	Bed height to bed diameter ratio [m/m]	Naik m	ik model	
IsoP	Isopropyl alcohol			
EtOH	Ethanol	е	The extract/solver	
CC	Curcuminoids	<i>e</i>	The extract/solver	
ρ_{Ap}	Apparent bed density [kg/m ³]		extraction	
VB	Extractor volume [m ³]	k	The adjustable par	
Dp	Particle diameter [m]	t	The extraction tim	
S^2	Mean square deviation			
OEC	Overall extraction curves	Goto m	o model	
CER	Constant extraction rate			
FER	Falling extraction rate	φ	Mass transfer coef	
Y _{CER}	The solute mass ratio in the fluid phase at the extractor	K	Adsorption equilib	
	exit in the CER period	β	Porosity of solid [
M _{CER}	Extraction rate in the CER period	X ₀	The maximum ext	
DF	Diffusional region	ρ_s	True density of so	
		Q_{CO2}	Mass flow rate of	
Crank model		ρ_{CO2}	Density of CO_2 [k	
		τ	Is the solvent resid	
$m_{\rm extract}$	The mass of extract [kg]	ε	Particles porosity	
X_0	Initial solute mass (kg)	ϕ	The dimensionless	
F	Mass of feed [kg]	k_p	Global mass trans	
n	The number of the series expansion	a_p	Specific surface ar	

SFE was studied by Chassagnez-Méndez et al. [3]. However, because of the low SFE yield in curcuminoids (CC), the addition of a co-solvent (ethanol or isopropyl alcohol) is necessary to improve their extraction [4,5,6]. The amount of CC extracted was found to be significantly affected when using ethanol as co-solvent [4]. Yadav et al. [7] studied the accelerated solvent extractor (ASE) of curcuminoids at low pressure, and the maximum yield and purity were obtained using ethanol and acetone, respectively. Braga et al. [5] and Braga and Meireles [6]. studied the influence of the pressurized ethanol, isopropyl alcohol, and their mixture (1:1 v/v) on the recovery of CC and concluded that the best co-solvent was the mixture ethanol:isopropyl alcohol (1:1 v/v), at 30 MPa and 303 K. The use of co-solvents in the SFE process has been widely reported, and the solubility of a solute and the selectivity of CO₂ towards that solute can be modified by the addition of a co-solvent. Additionally, the use of co-solvent decreases processing costs [8]. Osorio-Tobón et al. [9] studied the pressurized liquid extraction (PLE) with ethanol and the scale-up of curcuminoids extraction using the extractor volumes from 0.05 up to 0.5 m³; they also studied the cost of manufacturing for the turmeric extraction process.

The successful design of SFE processes requires studies on the solubility, kinetics and mass transfer data for solutes in supercritical fluids. Several studies have reported mass transfer models to describe the kinetic profile of SFE [10,8,11-17]. Cocero and García [11,12] studied several mathematical models to represent the SFE of lipids from sunflower. Because the experimental data showed a sigmoidal shape, the overall extraction curve (OEC) was modelled by the desorption model [11]. Two adjustable parameters (desorption equilibrium and mass transfer coefficient in the fluid phase) were considered. Cocero and García [12] evaluated three shortcut methods and concluded that the Fast Adjusting Method (FAM) predicted properly the OEC. This model is similar to the empirical model, but the difference is that it considers two adjustable parameters: *n* and *k*, where *n* is directly related with the curve, sigmoidal for n = 1. Zancan et al. [13] used the Sovová [18] model to represent the OEC from ginger rhizomes. Mantell et al. [14] proposed a penetration model and concluded that the controlling stage of the process was the internal diffusion. From these studies it can be concluded that the effect of the temperature, pressure, flow rate and

τ	The extraction time [s]				
r	The particle radius [m]				
Naik ma	odel				
е	The extract/solvent ratio [kg/kg]				
<i>e</i>	The extract/solvent ratio [kg/kg] in the infinite time of extraction				
k	The adjustable parameter [-]				
t	The extraction time [s]				
Goto ma	Goto model				
ф	Mass transfer coefficient [-]				
K	Adsorption equilibrium constant [-]				
β	Porosity of solid [-]				
X ₀	The maximum extract obtained in the solid (kg/kg)				
ρ _s	True density of solid $[kg/m^3]$				
Q_{CO2}	Mass flow rate of supercritical CO_2 [kg/s]				
Pco ₂	Density of CO_2 [kg/m ³]				
τ	Is the solvent residence time in the extraction cell [s]				
ε	Particles porosity [-]				
ϕ	The dimensionless mass transfer coefficient				
k_n	Global mass transfer coefficient [1/s]				
a_p	Specific surface area [1/m]				
-					

coefficient [m²/s]

co-solvent (percentage and type) on the kinetics of the SFE process can be complex.

Some mass transfer models have been used to fit data also obtained with co-solvents, mainly using the Sovová's model [19,20,21]. However, models applied to processes using co-solvents are scarce. Cygnarowicz-Provost et al. [8] used the Lee et al. [22] model to represent the OEC from fungal lipids. The mass transfer coefficient was the only adjustable parameter. These authors showed that the mass transfer coefficients with pure CO_2 and CO_2 + 10% of ethanol, can be represented by the same equation as a function of the Reynolds number. In this case, the mass transfer coefficients increased with increasing flow rate. Nevertheless, for extraction processes design purposes, information related with the kinetics and the mass transfer of the process is essential to predict the bed extract depletion. Thus, the objective of this work was to extend the previous studies, by applying four mathematical models to evaluate the OEC's obtained under the influence of different process variables that included pressure, co-solvent type, flow rate, co-solvent percentage and bed heights to bed diameter (H_B/D_B) ratio.

2. Materials and methods

The raw material and the SFE unit that were used to obtain the overall extraction kinetic curves modelled in the present work were previously described in Braga et al. [5] and Braga and Meireles [6]. Particles sizes 24, 32, and 48 mesh were selected for the assays, based on previous works considering the higher mass transfer into the fixed extraction bed. The mean particle diameter (D_p) was estimated using the procedure recommended by ASAE [23]. The real or true density (ρ_s) of the particles was measured by helium gas pycnometry (Multivolume Pycnometer Micromeritics, model 1305, Georgia, USA). The bed bulk density (ρ_{Ap}) was calculated using the extractor volume (V_B) and the mass of feed (F). The bed particles porosity (ε) was calculated using the apparent bed density and the particles real density, as an approximation, with the hypotheses of these particles do not have pores.

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