

# Supercritical CO<sub>2</sub> Extraction of Curcumins and Essential Oil from the Rhizomes of Turmeric (*Curcuma longa* L.)

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Turmeric rhizomes were extracted with supercritical CO<sub>2</sub> and supercritical CO<sub>2</sub> + ethanol. Extraction experiments were carried out at pressures of 25 and 30 MPa and temperatures of 313 and 318 K. The influence of the drying temperature of the raw material on the extraction yield and curcuminoids profile was evaluated. The higher content of curcuminoids in the extracts was obtained by supercritical fluid extraction from rhizomes dried at 343 K using CO<sub>2</sub> + ethanol. The identification of curcuminoids in both the extract and the residual solid was performed by both spectrophotometry and HPLC. The composition of the essential oil was determined by gas chromatographic mass spectrometry. A mathematical model was used to describe the overall extraction curves. The mass transfer inside the solid matrix was described by a linear first-order desorption model, whereas the transfer in the fluid phase was described by a convective mass-transfer model. The mathematical model fitted well the experimental data.

## Introduction.

The rhizome of turmeric (*Curcuma longa* L.) is an important source of a yellow natural pigment. The compounds responsible for the pigment yellow color are the curcumin [1,7-bis(4-hydroxy-3-methoxyfenil)-1,6-heptadiene-3,5-dione] and two curcuminoids [demethoxycurcumin (DMC) and bis(demethoxy)curcumin (BD-MC)].<sup>1,2</sup> These pigments are largely used in the food industry as substitutes for synthetic dyes such as tartrazin. In addition, pharmacological investigations have shown that curcumin acts as a bactericide and antiinflammatory agent.<sup>3</sup> Curcumin is a yellow-orange crystalline powder insoluble in water, poorly soluble in hydrocarbon solvents, and soluble in alcohols. The extraction of oleoresin from a dried turmeric powder has been performed at laboratory and pilot scale by Soxhlet extraction and cold and mild percolation using ethylene dichloride, ethanol, or acetone as the solvent.<sup>4–6</sup> In those works, the extraction yields varied from 3.20 to 9.95 wt % for the Soxhlet method and from 3.60 to 10.42 wt % for the percolation method. Despite the relatively high extraction yields, several problems contribute to the drawback of solvent extraction using ethylene dichloride, acetone, and alcohol to obtain curcuminoids from turmeric. Acetone presents problems of flammability and high recovery costs, ethylene dichloride produces traces of high boiling fractions in the extracts, creating off-flavor problems, and extraction with ethanol needs a water-washing step to remove the solvent.

In the past few years, numerous applications of supercritical fluid extraction (SCFE) with CO<sub>2</sub> from

different vegetable materials have been reported in the literature including herbaceous and spice materials.<sup>7–9</sup> Recently, there has been a great interest in SCFE of pigments, among these the carotenoids from different raw materials such as alfalfa,<sup>10</sup> sweet potatoes,<sup>11</sup> buriti,<sup>12</sup> tucumã,<sup>13</sup> and the residue of the mechanical expulsion of palm oil.<sup>14,15</sup>

The objective of this work was to study the kinetics of SCFE from turmeric rhizomes. Because of the large content of water, the rhizomes must be dried prior to SCFE. Also, the turmeric extracts are formed by a mixture of polar and nonpolar substances. Therefore, the specific objective of the present work was to analyze the influence of the rhizomes' drying temperature and the addition of ethanol (EtOH) as a cosolvent on the extraction yield and product quality for the turmeric oleoresin obtained by SCFE. In addition, a mass-transfer model was applied to describe the overall extraction curves.

## Materials and Methods

**Raw Material Pretreatment and Characterization.** The rhizomes of turmeric (*Curcuma longa* L.) were obtained from an experimental plantation of INCRA (Altamira, Pará, Brazil). Carbon dioxide (99.99 wt % pure) was provided by White Martins S/A.

The rhizomes of turmeric were washed with water to remove the remaining sand and to reduce the microbial load. The cleaned rhizomes were cut (0.6–0.9 cm) and dried in an oven with air circulation (Fabbe, Piracicaba, Brazil; model 170) at 343 and 378 K for 24 h. The dried rhizomes were grounded in a comminuting mill (Framo-Gerätetechnik, Germany; model A70).

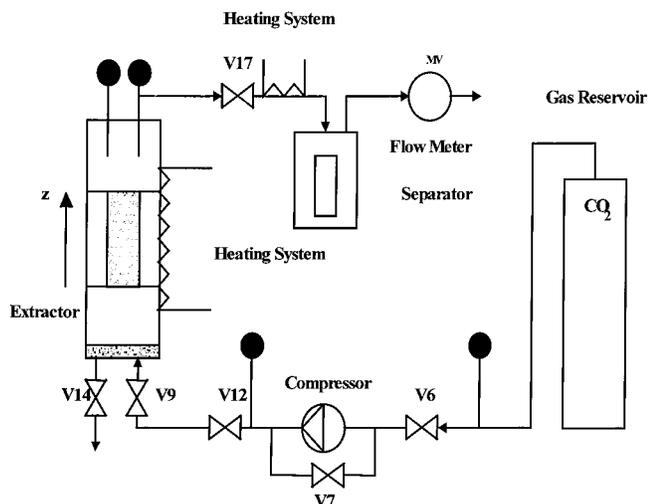
To compute the mean particle diameter, the dried and grounded solid material was sifted in a sieve shaker (Bertel, Caieiras, Brazil; model N800) with sieves of different sizes (Tyler; 48, 65, 80, 100, and 150 mesh). The dried material was packed in plastic bags and kept in a dark environment at 298 K to avoid photodegra-

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**Figure 1.** Experimental setup.

dation of the curcuminoids, to be used later in the extraction experiments.

The density of the dried solid was determined by the sand picnometry technique,<sup>16</sup> the mean particle diameter ( $D_{\text{Sauter}}$ ) was calculated as suggested by Massarani,<sup>17</sup> and the content of the curcuminoids (expressed in terms of curcumin) was calculated by the method proposed by Takahashi.<sup>18</sup>

**Experimental Setup.** A schematic diagram of the extraction unit used in this work is shown in Figure 1. It consists basically of one extractor (Metalwerksatt, TUHH) of 1000 mL, a diaphragm-type compressor (Andreas Hofer, Mülheim, Germany; model MKZ 120-50), one separator (Mechanical Workshop, UFPa) of 50 mL, a thermostatic bath (Haake Mess-Technik GmbH, Karlsruhe, Germany; model N3), a carbon dioxide reservoir, a gas flowmeter (Bopp Reuter Mess-Technik GmbH, Maisach, Germany), and a control unit that displays the temperature and pressure inside the extractor. To reduce the size of the extractor, a device constructed of stainless steel was placed inside it, as indicated in Figure 1.

**Experimental Procedure.** Carbon dioxide was delivered at the required pressure by the membrane compressor and passed through a porous plate in order to ensure a homogeneous flow of the solvent along the fixed bed constructed. For the experiments with supercritical  $\text{CO}_2$ , a fixed bed of 19.80 cm height and 1.76 cm internal diameter was used, while for the experiments with supercritical  $\text{CO}_2 + \text{EtOH}$ , the fixed-bed dimensions were 4.00 cm of height and 2.54 cm of internal diameter. The pressure inside the extractor was measured by a Bourdon-type gauge ( $0-400 \pm 5$  bar; Wika, Germany; model DIN.S), and the temperature was monitored by a thermocouple (NiCr/Ni). After extraction the solute/solvent mixture was expanded in the separator. The extracts condensed in the separator were weighed using a semianalytical balance (Gehaka, São Paulo, Brazil; model 4400). The expanded carbon dioxide passed through a gas flowmeter containing a thermometer and was released to the atmosphere. For the experiments using ethanol as the cosolvent, the supercritical  $\text{CO}_2$  has passed through a high-pressure autoclave containing ethanol before entering the extractor. It was not possible to measure the EtOH content in the gaseous phase. EtOH was removed from the condensate

(oleoresin + EtOH) using a rotary evaporator (Quimis, Brazil; model 3442).

**Chemical Quantification of the Curcuminoids in the Oleoresin by Spectrophotometry.** To quantify the curcuminoids in the extracts, the oleoresin was diluted in EtOH (PA, Quimis, Diadema, Brazil) and the samples were analyzed in a spectrophotometer (GBC, model UV/VVIS 916) at 425 nm. The absorbance was measured and the content of curcuminoids in the oleoresin expressed in weight percent,<sup>18</sup>

**Chemical Quantification of the Curcuminoids in the Residual Solid by Spectrophotometry.** The chemical quantification of the curcuminoids (expressed in terms of curcumin) present in the residual solid was determined by Soxhlet extraction using EtOH as the solvent (PA, Quimis, Diadema, Brazil) for 2 h. The solvent was evaporated in a vacuum rotary evaporator (Quimis, Diadema, Brazil; model 3442). The quantification of the curcuminoids was performed using the procedure described previously.

**Chemical Quantification of the Curcuminoids by HPLC.** The chemical quantification of curcumin and the two curcuminoids [DMC and BDMC] in the oleoresin was performed by high-pressure liquid chromatography (HPLC) using the procedure that follows.<sup>19</sup> The samples (1.0–9.0 mg) were diluted in a standard solution of acetic acid (1.0 mg/mL) with the pH adjusted to 2.88, prepared in the proportion of 55:45 (v/v) with acetonitrile (chromatographic grade; Science) and deionized water filtered using membranes of nylon with pores of 0.45 (Supelco). Afterward, the mobile phase at a rate of 1 mL/min was injected in the HPLC (Shimadzu model LC-10AD) equipped with an UV-vis detector (Shimadzu model SPD-10A), a manual splitting system (Shimadzu model SV-10AM), a stationary phase column (Shimadzu model Hypersil ODS- $\text{C}_{18}$ ) of 250 nm  $\times$  4.5 nm, and a communication system (Shimadzu model CBM-10A) coupled to a personal computer and control software (Shimadzu class LC10). The detector was adjusted at 425 nm and 298 K. The concentration of curcumin and the two curcuminoids [DMC and BDMC] in the oleoresin expressed in micrograms per milliliter was computed using calibration curves, constructed with standard solutions of curcuminoids (PA, Merck, Darmstadt, Germany).

**Chemical Identification of the Essential Oil in the Oleoresin of Turmeric by Gas Chromatographic Mass Spectrometry (GCMS).** The chemical quantification of the essential oil was performed by GCMS using a standard procedure.<sup>19</sup> The SCFE extracts were submitted to hydrodistillation for 2 h. The essential oil, extracted with pentane, was injected in the GCMS system consisting of a gas chromatograph (Varian model 3400) coupled to a mass spectrometer (Finnigan model INCOS-XL). A fused silica capillary column DB-5 (J&W Scientific; 30 m  $\times$  0.245 mm, 0.25  $\mu\text{m}$  coated with 5% phenylmethylpolysiloxane) was used. The carrier gas was helium (32 cm/s). The temperature program was as follows: oven temperature, 333–513 K (4 K/min); detector temperature, 493 K. The mass spectrophotometer was operated at 453 K. The electron impact technique (70 eV) was used. The compounds were identified by comparison of their mass spectrum and retention indices with those of standard substances available in the reference library of the GCMS system.<sup>20,21</sup>

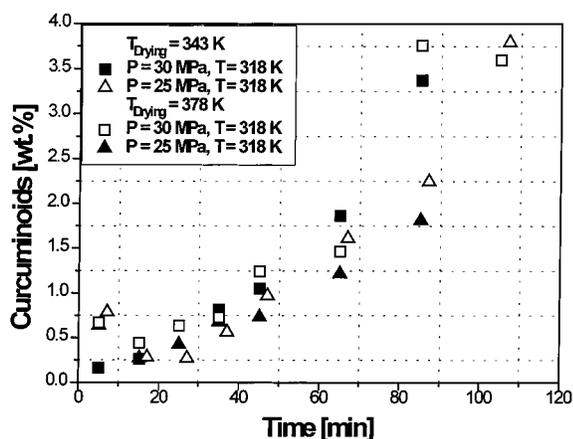
**Table 1. Characteristics of the Fixed Bed Used for SCFE**

	drying temp of raw material	
	343 K	378 K
bed density [g/cm <sup>3</sup> ]	0.73 ± 0.03	0.75 ± 0.05
porosity	0.44 ± 0.02	0.43 ± 0.03
$D_{\text{Sauter}}$ [ $\mu\text{m}$ ]	207	214
$V_{\text{fixed bed I}}$ [cm <sup>3</sup> ]	27.37	26.51
$V_{\text{fixed bed II}}$ [cm <sup>3</sup> ]	27.37	26.51

**Table 2. Process Parameters and Operating Conditions for the Extraction of Turmeric's Oleoresin (*Curcuma Longa* L.) with Supercritical CO<sub>2</sub> at 318 K and Supercritical CO<sub>2</sub> Using EtOH as the Cosolvent at 318 K**

solvent	pressure [MPa]	temp [K]	total extraction time [min]	drying temp [K]	$m_{\text{solvent}}$ [g/min]	yield <sup>a</sup> [wt %]
CO <sub>2</sub>	25	318	107	343	14.65	5.95
CO <sub>2</sub>	30	318	85	343	11.21	6.51
CO <sub>2</sub>	25	318	85	378	11.33	4.50
CO <sub>2</sub>	30	318	105	378	13.27	5.28
CO <sub>2</sub> + EtOH	25	318	95	343	9.75	13.42
CO <sub>2</sub> + EtOH	30	318	95	343	6.54	22.58

<sup>a</sup> Mass of extracted oleoresin/mass of dried feed.

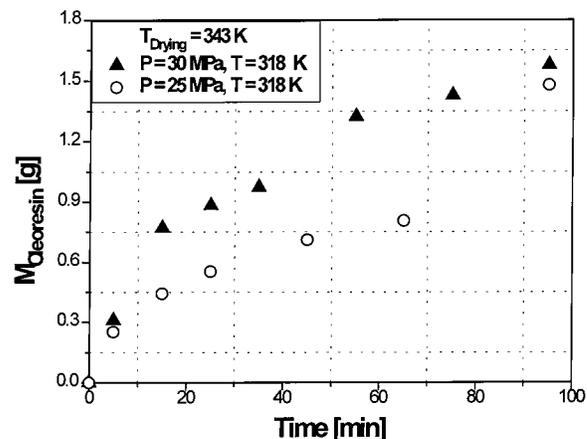
**Figure 2.** Experimental profiles of curcuminoids obtained by SCFE with CO<sub>2</sub>.

## Results and Discussion

The fixed-bed characteristics are given in Table 1. The amounts of curcumin in the dried rhizomes were  $8.9 \pm 0.2$  and  $6.4 \pm 0.1$  wt % for drying temperatures of 343 and 378 K, respectively.

Table 2 shows the operational variables and the extraction yields (mass of extract/mass of dried feed). For SCFE using CO<sub>2</sub>, the maximum yield and the minimum solvent consumption were achieved at 30 MPa and 318 K for rhizomes dried at 343 K. The addition of EtOH as the cosolvent on the second series of experiments increased the extraction yield and decreased the solvent consumption. Despite this, the use of a cosolvent requires another separation step. The yields for SCFE of turmeric oleoresin with CO<sub>2</sub> were close to those obtained by Soxhlet extraction. SCFE with CO<sub>2</sub> + EtOH increased the yields, as compared to the Soxhlet extraction and to the percolation method.<sup>4-6</sup>

The curcuminoids (expressed in terms of curcumin) profile is shown in Figure 2. The figure illustrates the effects of the drying temperature on the curcuminoids profile. At 30 MPa as well as at 25 MPa up to 50 min, the effect of the drying temperature is small. Afterward, as the amount of curcuminoids extracted increases, the effect of the drying temperature is more pronounced. At this stage it is clear that drying the rhizomes at 378

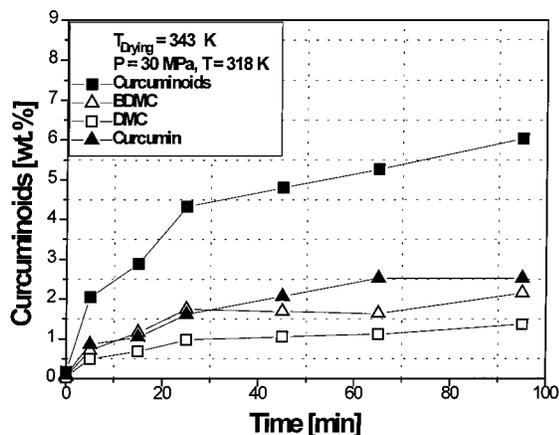
**Figure 3.** Overall extraction curves for SCFE from turmeric rhizomes obtained with CO<sub>2</sub> + EtOH at 318 K and with pressures of 25 and 30 MPa.

K decreased the content of pigments in the turmeric extract. This fact may be attributed to the larger loss of oleoresin by volatilization that occurs at the higher drying temperature as compared to the lower one. In addition to that, for all experiments the content of curcuminoids increased during the course of extraction. This phenomenon may indicate that the equilibrium solubility was not reached, while a great amount of curcuminoids still remained in the solid matrix. It was observed that the effect of both the pressure and the drying pretreatment on the concentration became significant as the extraction time increased. The higher content of curcuminoids in the oleoresin was achieved at 30 MPa for rhizomes dried at 343 K.

Figure 3 shows the overall extraction curves for SCFE using CO<sub>2</sub> + EtOH, for rhizomes dried at 343 K. The behavior of the overall extraction curve at 30 MPa and 318 K has the traditional shape.<sup>8,12,15</sup> At 25 MPa, the shape of the overall extraction curve is unusual but the total yield at both pressures was approximately the same. In both cases, the addition of EtOH increased the total yield (Table 2). The polarity of the mixture CO<sub>2</sub> + EtOH permits the solubilization of more polar substances present in the turmeric rhizomes than that of CO<sub>2</sub> can permit by itself.

The content of curcumin, DMC, BDMC, and curcuminoids (total) in the turmeric oleoresin, obtained by SCFE with CO<sub>2</sub> + EtOH at 30 MPa and 318 K from rhizomes dried at 343 K, is shown in Figure 4. The figure shows that the content of pigments increased during the extraction. When the content of pigments originally present in the vegetable material is compared with the amount in the extract, it can be observed that most of the curcuminoids still remained in the solid matrix. Therefore, to increase the curcuminoids extraction, another technological solution is required besides the use of EtOH as the cosolvent.

Table 3 shows the chemical composition of the essential oil for the extract obtained using CO<sub>2</sub> + EtOH at 25 MPa and 318 K from rhizomes dried at 343 K. Some literature data for the extract obtained by solvent extraction are also shown in Table 3.<sup>23,24</sup> It can be observed that the CO<sub>2</sub> + EtOH extract is formed by a complex multicomponent mixture. For the substances reported in the literature, their amounts in the SCFE were always greater than the amount in the solvent extract.



**Figure 4.** Profiles of curcumin, DMC, BDMC, and curcuminoids (total) at 30 MPa and 318 K.

**Table 3. Composition of the Turmeric Essential Oil Obtained by SCFE with CO<sub>2</sub> + EtOH along with Literature Information for the Extract Obtained with Organic Solvent**

compound	retention index	composition [wt %]	ref 24	ref 25
α-pinene	937	0.1	0.03–0.1	
mirocene	989	0.2		
α-felandrene	1005	4.1		
α-terpinene	1017	0.1		
p-cimene	1024	1.5		
limonene	1029	0.4		
1,8-cineole	1032	4.0	0.07–0.13	0.13–1.14
γ-terpinene	1060	0.2		
terpinolene	1087	1.3		0.10–4.16
p-cimonene	1088	0.04		
4-terpinol	1180	0.2		
8-p-cimanol	1184	0.2		
α-terpineol	1191	0.4		
timol	1292	0.2		
carvacrol	1300	0.03		
α-cis-bergatomena	1413	0.1		
cariofilene	1420	0.9		0.15–4.5
α-humulene	1457	0.6		
γ-curcumene	1481	0.2		
ar-curcumene	1485	3.6	2.6–6.5	1.42–4.80
α-zingiberene	1497	6.4	3.2–6.0	0.85–14.07
β-bisabolene	1510	1.7		0.29–2.20
β-sesquifelandrene	1523	7.7		1.47–13.34
β-germacene	1556	0.2		
ar-turmerol	1581	1.5		
ar-turmerone	1670	15.5	7.68–20.30	7.71–30.71
(Z)-γ-atlantona	1688	20.3		
(E)-γ-atlantona	1706	15.6		
(6S,7R)-bisabolene	1743	0.3		
(E)-α-atlantona	1778	0.6		

**Mathematical Modeling of SCFE.** Several models have been used in the literature to describe the overall extraction curves. Among them, the models discussed by Reverchon et al.,<sup>9</sup> Sovová,<sup>24</sup> and Tan and Liou<sup>25</sup> can be mentioned. The simplest one is the single-parameter model used to describe the desorption at supercritical conditions from activated carbon by Tan and Liou.<sup>25</sup> This model was chosen to describe SCFE from turmeric rhizomes because it can easily be incorporated into any simulating routine.

The single-parameter model applied to SCFE from turmeric rhizomes considers that the mass transfer in the fluid phase can be described by a nonstationary convective model. The model has a term describing the mass transfer at the solid–fluid boundary (the solute transfer rate leaving the solid boundary is equal to the solute transfer rate entering the solvent phase), without considering the axial dispersion through the fixed bed.

It considers also that the extraction rate of oleoresin in the solid phase is proportional to its concentration (linear first-order desorption), in accordance with the desorption model reported in the literature.<sup>28</sup> The phenomenon in the solid phase can be interpreted as a transport process with instantaneous equilibrium, based on the variations of solute chemical composition, on the content of the vegetable material, and on a small mass-transfer resistance. The mass balance for an element of the bed written in terms of mass ratio is given by

mass balance for the bulk fluid phase

$$\frac{\partial Y(t,z)}{\partial t} + \langle U \rangle \frac{\partial Y(t,z)}{\partial z} = - \frac{1 - \epsilon}{\epsilon} \frac{\rho_S}{\rho_f} \frac{\partial X(t)}{\partial t} \quad (1)$$

initial and boundary conditions

$$\text{I. } Y(t,z) = 0, \quad t = 0 \quad (2)$$

$$\text{II. } Y(t,z) = 0, \quad z = 0 \quad (3)$$

mass balance in the solid phase

$$(1 - \epsilon) \rho_S \frac{\partial X(t)}{\partial t} = -KX(t) \quad (4)$$

initial condition

$$\text{I. } X(t) = X_0, \quad t = 0 \quad (5)$$

The solute mass ratio in the fluid phase at the fixed-bed outlet can be obtained by simultaneously solving eqs 1 and 2 with the boundary and initial conditions given by eqs 3–5, resulting in

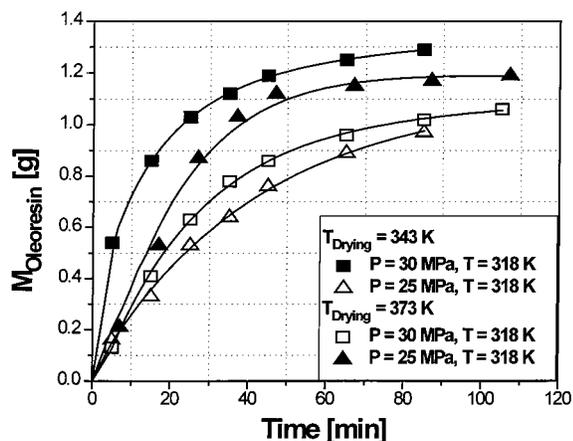
$$Y(z=L,t) = \frac{1 - \epsilon}{\epsilon} \frac{\rho_S}{\rho_f} X_0 \left\{ \exp \left[ -K \left( t - \frac{L}{\langle U \rangle} \right) \right] \exp(-Kt) \right\} \quad (6)$$

The overall extraction curve (mass of oleoresin extracted as a function of the extraction time) is given by

$$M_{\text{oleoresin}} = \int_0^t \dot{m}_{\text{solvent}} Y(z=L,t) dt \quad (7)$$

where  $Y$  is the solute mass ratio in the fluid phase ( $\text{g}_{\text{solute}}/\text{kg}_{\text{CO}_2}$ ),  $X$  is the solute mass ratio in the solid phase ( $\text{g}_{\text{solute}}/\text{kg}_{\text{solid}}$ ),  $\langle U \rangle$  is the interstitial velocity (m/s),  $L$  is the fixed-bed length (m),  $K$  is the first-order desorption constant ( $\text{s}^{-1}$ ),  $\dot{m}_{\text{solvent}}$  is the solvent mass flow rate (kg/s),  $\rho_S$  is the solid-phase density ( $\text{kg}/\text{m}^3$ ),  $\rho_f$  is the fluid-phase density ( $\text{kg}/\text{m}^3$ ), and  $\epsilon$  is the fixed-bed porosity.

Tan and Liou<sup>25</sup> determined that the desorption constant is a function of the fluid-phase density. It is well-known that for SCFE from solid substratum the solubility of the extract in the solvent is a function of the solvent density. Using solubility data for the turmeric extract in CO<sub>2</sub>, it would be possible to estimate the desorption constant. Unfortunately, there are no data for the solubility of the turmeric oleoresin in supercritical CO<sub>2</sub>. Therefore, the alternative was to treat the constant of desorption ( $K$ ) as an empirical parameter. Table 4 shows  $K$  obtained by fitting the experimental overall extraction curves to eq 7. The value of  $K$  increased with solvent density, as would be expected because the parameter is closely related to the solubility of the solute in the solvent. The physical mechanism behind the Tan and Liou model<sup>21</sup> assumes that the



**Figure 5.** Experimental and calculated overall extraction curves for SCFE with CO<sub>2</sub> from turmeric rhizomes.

**Table 4. Constant of Desorption  $K$  and Mass Ratio of the Solute in the Fluid Phase at the End of SCFE**

pressure [MPa]	drying temp [K]	$\rho_{\text{CO}_2}$ [kg/m <sup>3</sup> ]	$K$ [min <sup>-1</sup> ]
25	343	737	0.0406
30	343	789	0.0857
25	378	567	0.0258
30	378	643	0.0335

desorption process takes place at the solid–fluid interface and that it can be described using a first-order reaction kinetics. Therefore, it should be used with systems with relatively high desorption constants, that is, high mass-transfer rates at the solid–fluid interface. However, this is not the case for SCFE from turmeric rhizomes. Figure 5 shows that the experimental overall extraction curves agree well with the calculated ones. Araújo et al.<sup>26</sup> have used the same model to successfully describe SCFE from *Guilielma speciosa*, a fruit of a palm tree from the Northern region of Brazil.

## Conclusions

The extraction yields were affected by the temperature used for drying the rhizomes of turmeric. Drying the rhizomes at 378 K resulted in a larger loss of volatile material, resulting in lower total yields as compared to drying at 343 K. Although the use of EtOH as the cosolvent increased the yield, because it favored the solubilization of the more polar substances present in the rhizomes, part of the curcuminoids remained in the solid matrix. Therefore, additional studies are required in order to develop SCFE applied to turmeric processing. Although the desorption model assumptions were not entirely valid for the system turmeric rhizomes/CO<sub>2</sub>, the overall extraction curves were well described by the model, using the constant of desorption estimated from the experimental data.

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