

Supercritical Fluid Extraction of β -Carotene from Crude Palm Oil Using CO₂ in a Bubbler Extractor: Mass Transfer Study

Davarnejad, Reza*⁺

*Department of Chemical Engineering, Faculty of Engineering, Arak University,
P.O. Box 38156-8-8349 Arak, I.R. IRAN*

Niza, Norazlila Mohammad

*School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia,
14300 Nibong Tebal, Penang, MALYSIA*

Arpanahzadeh, Shahrzad; Zakeri, Melika

*Department of Chemical Engineering, Faculty of Engineering, Arak University,
P.O. Box 38156-8-8349 Arak, I.R. IRAN*

ABSTRACT: *In this study, the diffusivity and volumetric mass transfer coefficients for the extraction of β -carotene from crude palm oil using CO₂ solvent in supercritical fluid were determined in the pressure range of 7.5-17.5 MPa and temperature range of 80-120 °C. For this purpose, a statistical method was applied in order to minimize the number of experiments required. The volumetric mass transfer coefficient for the extraction of β -carotene was then correlated as a function of pressure, temperature and extraction time in order to study the effect of each of these variables on the coefficient. The experimental results showed that the maximum volumetric mass transfer coefficient was around $2.486 \times 10^{-2} \text{ s}^{-1}$ at pressure of 7.5 MPa, temperature of 100 °C and extraction time of 1 hour. The minimum volumetric mass transfer coefficient was $0.046 \times 10^{-2} \text{ s}^{-1}$ at pressure of 17.5 MPa, temperature of 120 °C and extraction time of 5 hours. The optimum volumetric mass transfer coefficient was statistically obtained around $6.700 \times 10^{-3} \text{ s}^{-1}$ at pressure of 17.7 MPa, temperature of 100.5 °C and extraction time of 3.9 hours.*

KEY WORDS: *β -carotene, Diffusion, Extraction, Mass transfer, Optimization, Supercritical fluid.*

INTRODUCTION

Crude Palm Oil (CPO) is one of Malaysia's main commodity products. Its characteristics were illustrated in Table 1.

CPO also has a significant β -carotene weight percentage, which is about 0.027%-0.038%, and is a precursor of vitamin A in human and animal metabolism [1].

* To whom correspondence should be addressed.

+ E-mail: r-davarnejad@araku.ac.ir

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β -carotene, an orange color lipid precursor of vitamin A is usually added to food products in order to give a uniform coloring. Although many vegetables are rich in carotenoids such as β -carotene, however a large portion of commercially available β -carotene is still synthetically produced due to the difficulty in extracting β -carotene from natural products [2]. For instance, the carotenoids percentage in palm oil is around 500-700 mg/kg with a maximum carotene isomer of 54.4 % [3]. One possible method to extract out this valuable component economically is via supercritical fluid extraction such as carbon dioxide.

Supercritical fluid extraction of β -carotene from Crude Palm Oil (CPO) using carbon dioxide was already studied by *Davarnejad et al.* [4]. They showed that the maximum yield of the β -carotene (1.741×10^{-2} %) was experimentally obtained at a pressure of 7.5 MPa, temperature of 120 °C and extraction time of 1 hour.

However, in order to further increase the extraction yield of β -carotene, understanding the mass transfer properties of the process might be a crucial factor. A common method to study the mass transfer properties is by identifying suitable mathematical equations based on experimental results that are carried out with varying process parameters such as temperature, pressure, solvent velocity, particle size and physical properties of supercritical fluids [5-7]. Mass transfer extraction process of β -carotene from carrot using CO₂ and N₂O was studied by *Subra et al.* [5]. They concluded that the mass transfer coefficient was around 2.5×10^{-8} m/s with the maximum value achieved at the following conditions; temperature of 57 °C, pressure of 250 bar, solvent density of 0.8 g/cm³, flow rate of 0.73 g/min and particle size of 0.5 mm. They also concluded that increasing the particle size increased the mass transfer coefficient. Furthermore, it was observed that the mass transfer coefficient in N₂O solvent was higher than the mass transfer coefficient in CO₂ solvent at the same operating conditions.

Shi et al. have correlated mass transfer coefficient in the extraction of plant oil in a fixed bed for supercritical carbon dioxide [6]. According to this study, the estimation result based on the data of *Puiggene et al.* was revised as $Sh' = 0.422(Re^{0.580} Sc^{0.3074})$ when $10 < Re < 100$. It shows consistent kinetic behavior in this

Table 1: Malaysia CPO characteristics [1].

Components	Weight percentage
Saturated fatty acids	45.5
Monounsaturated fatty acids	39.6
Carotenoids	0.05-0.07
Tocopherols and tocotrienols	0.06-0.1
Sterols	0.0326-0.0527
Phospholipids	0.0005-0.0130
Triterpene alcohol	0.004-0.008
Methyl sterols	0.004-0.008
Squalene	0.02-0.05
Aliphatic alcohols	0.01-0.02
Aliphatic hydrocarbons	0.005

region according to their data and it is also validated by other data reported in the literature. The estimation also shows that it is acceptable to account the exponent of exponential term of Sc number as 1/3. Other mass transfer studies on the extraction of valuable components from solid matrixes using supercritical CO₂ are by *de França et al.* [7] and *King et al.* [8]. *de França et al.* studied the yield of carotene extraction from buriti (that is a tropical fruit) while *King et al.* studied and modeled the extraction from various natural products.

Tan & Liou measured the axial dispersion coefficients under supercritical conditions by injection of methane in the supercritical carbon dioxide. They correlated the axial dispersion with the Reynolds and Schmidt numbers. Furthermore, they found that the correlations based on the Peclet, Reynolds and Schmidt dimensionless groups could not predict the mass transfer, properly [9].

In this study, β -carotene mass transfer properties such as diffusivity and volumetric mass transfer coefficients from crude palm oil in supercritical fluid extraction process using CO₂ solvent were determined and statistically modeled. Further, the effects of various process variables such as extraction time, temperature and pressure on this factor were investigated and the optimum volumetric mass transfer coefficient within the operating range studied was statistically obtained.

For this purpose, experimental results [yield of β -carotene extraction from CPO (g extracted β -carotene/g CPO) as function of pressure, temperature and extraction time] which were already published [4] with some general mass transfer equations in supercritical fluid extraction process [10] were applied. Another necessary parameter that is required for this study is the β -carotene equilibrium percentages in the extracted phase which were also interpolated from the literature [1].

THEORITICAL SECTION

Mass Transfer Study

The data used in this study is based on the extraction of β -carotene from crude palm oil using CO_2 as reported in our previous study [4]. Although Catchpole has comprehensively studied the mass transfer phenomenon in a packed-bed extractor and has derived relevant equations [11], however, the equations might not be suitable for this study due to the nature of palm oil. Therefore, in this study, more common equations that are normally used in mass transfer study were applied to obtain two significant mass transfer properties namely, diffusivity and volumetric mass transfer coefficient. A substantial body of data now exists for the diffusion of heavy solutes in near-critical fluids as tabulated by King & Bott [10]. Based on the survey of these data, Catchpole has generated the following predictive equations:

For self diffusion:

$$D_{11} = \frac{D_c T_r (\rho_r^{1/3} - 0.4358\rho_r)}{0.5642\rho_r} \quad 0.4 \leq \rho_r \leq 2.5 \quad (1)$$

$$D_{11} = \frac{D_c T_r}{\rho_r} \quad 0.4 > \rho_r \quad (2)$$

D_c is the self diffusion coefficient of the solvent at the critical point. T_r and ρ_r are the reduced temperature and density respectively.

For binary diffusion:

$$D_{12} = 2^{\frac{3}{2}} \frac{D_{11} R}{X} \quad \rho_r < 2.5 \quad (3)$$

where,

$$X = \frac{[1 + (V_{c2} - V_{c1})^{1/3}]^2}{(1 + \frac{M_1}{M_2})^2} \quad (4)$$

V_{c1} and V_{c2} are the critical volumes of components 1 and 2 and M_1 and M_2 are their molecular weights.

For cyclic solutes, R is given by:

$$R = 0.644X^{0.17} \quad 2 \leq X \leq 10 \quad (5)$$

For linear solutes,

$$R = 0.611X^{-2} - 0.145X + 1.32 \quad 2^{1.5} \leq X \leq 10 \quad (6)$$

$$R = 1 \quad 2.5 \leq X \quad (7)$$

The self diffusion coefficient at the critical point, D_c required in Eq. (1) can be obtained from literature [10]. Also, an estimate for D_c can be obtained from the critical properties of the fluid [12]:

$$D_c = 0.77 \times 10^{-9} \left[\frac{P_c^{1/2} V_c^{5/6}}{M^{1/2}} \right] \quad (8)$$

with P_c in atmospheres, V_c in $\text{cm}^3 \text{mol}$ and M in g/mol .

A better estimate can be obtained by modifying the Fuller-Schettler-Giddings [13] empirical diffusion coefficient to give D_{11} in terms of ρ_c assuming D_{12} is constant up to the critical point [14]:

$$D_c = \frac{4.300 \times 10^{-7} M^{1/2} T_c^{0.75}}{\sum_v^{2/3} \rho_c} \quad (9)$$

with M in g/mol , T_c in K , ρ_c in kg/m^3 . The related volumes (\sum_v) are given in the literature [14].

By applying Eq. (9), D_c for CO_2 was around $4.9370 \times 10^{-8} \text{ m}^2/\text{s}$. Furthermore, by integration from the equation $\dot{V}dC = k_f a (C^* - C) dv$ (where \dot{V} , dC , k_f , a , C and C^* are volumetric flow rates, the increase in solute concentration in the solvent stream as it flows through the volume element dv , mass transfer coefficient, mass transfer area, concentration of solute in the solvent within this element and the equilibrium value of C , respectively) from the bottom to the top of the extractor, concentration profile can be obtained [10]:

$$\frac{C_T}{C^*} = 1 - e^{(-k_f a \tau)} \quad (10)$$

Where τ is extractor residence time.

Assuming that the changes of feed volume during extraction process is negligible and the extractor residence time is equal to the extraction time (due to the use of a re-circulating pump which is able to circulate the gas phase continuously), β -carotene solubilities data

Table 2: Concentration of β carotene at extraction times 1, 3 and 5 h: temperature 80, 100 and 120 °C.

P (MPa)	$C_T \times 10^{-7}$ (mol/cm ³)	P (MPa)	$C_T \times 10^{-7}$ (mol/cm ³)	P (MPa)	$C_T \times 10^{-7}$ (mol/cm ³)
1 h and 80 °C		1 h and 100 °C		1 h and 120 °C	
17.5	2.004	17.5	2.147	17.5	2.289
12.5	2.524	12.5	1.784	12.5	2.422
7.5	2.459	7.5	1.277	7.5	2.556
3 h and 80 °C		3 h and 100 °C		3 h and 120 °C	
17.5	1.831	17.5	2.234	17.5	2.091
12.5	2.411	12.5	2.124	12.5	2.008
7.5	2.287	7.5	2.128	7.5	1.478
5 h and 80 °C		5 h and 100 °C		5 h and 120 °C	
17.5	1.599	17.5	1.470	17.5	1.341
12.5	1.895	12.5	1.466	12.5	1.697
7.5	2.242	7.5	1.463	7.5	2.052

can then be calculated using regular numerical solutions to the equations above [1]. Subsequently, using interpolation method, the β -carotene solubilities data were then determined as a function of temperature and pressure [1].

EXPERIMENTAL SECTION

Materials and method

According to the experimental work carried out by Davarnejad et al. [4], crude palm oil was purchased from United Oil Palm Industries Sdn. Bhd., Malaysia. CO₂ as solvent (99.99 %) was purchased from MOX Sdn. Bhd., Malaysia. β -carotene (96 %, Fluka), acetone (99.8 %, Baker) and n-hexane (99.8 %, Baker) were also used in the experimental analysis. The experimental process was conducted in a rate test supercritical fluid extraction unit which has been equipped with a bubbler extractor [4]. The extracted samples were analyzed by UV-Vis at wavelength of 450 nm to obtain β -carotene absorption. The experimental results were reported by Davarnejad et al. [4]. In the current paper, those data were converted to concentration (Table 2) to obtain mass transfer data.

RESULTS AND DISCUSSION

Diffusivity and Mass Transfer Coefficient

By applying Eqs. (1) to (10), the diffusivity and mass transfer coefficients for the extraction of β -carotene from crude palm oil using CO₂ were calculated. Initially, the diffusivity coefficients were calculated and are as listed in Table 3.

Subsequently, the diffusivity coefficients were used to calculate the volumetric mass transfer coefficients and are as listed in Table 4. It was observed that the calculated volumetric mass transfer coefficients data were the same (up to three decimal points) for the repeated runs as reported in Table 4. Furthermore, Table 4 showed that maximum volumetric mass transfer coefficient ($2.486 \times 10^{-2} \text{ s}^{-1}$) was obtained at the pressure of 7.5 MPa, temperature of 100 °C and extraction time of 1 hour while minimum volumetric mass transfer coefficient ($0.046 \times 10^{-2} \text{ s}^{-1}$) was obtained at the pressure of 17.5 MPa, temperature of 120 °C and extraction time of 5 hours.

Statistical analysis

The volumetric mass transfer coefficients data were then analyzed using Design Expert version 6.0.6 software to correlate volumetric mass transfer coefficient as a function of pressure, temperature and extraction time. For this purpose, Response Surface Method (RSM) coupled with Central Composite Design (CCD) was selected. Central Composite Design (CCD) was reported to be the most popular design for RSM because of its ability and suitability to fit quadratic surface which usually works well for process optimization [4]. Furthermore, the significance of the effect of each parameter on volumetric mass transfer coefficient can be identified using this software. Based on the data fed to the software, it was suggested that a quadratic model best fit the correlation between the volumetric mass transfer coefficient and

Table 3: Diffusivity coefficient at extraction time 1, 3 and 5 h: temperature 80, 100 and 120 °C.

P (MPa)	$D_{AB} \times 10^5$ (m ² /s)	P (MPa)	$D_{AB} \times 10^5$ (m ² /s)	P (MPa)	$D_{AB} \times 10^5$ (m ² /s)
1 h and 80 °C		1 h and 100 °C		1 h and 120 °C	
17.5	2.857	17.5	4.715×10^{-3}	17.5	3.951
12.5	5.235×10^{-3}	12.5	5.845×10^{-3}	12.5	3.572
7.5	4.178	7.5	9.741×10^{-3}	7.5	1.082×10^{-2}
3 h and 80 °C		3 h and 100 °C		3 h and 120 °C	
17.5	2.857	17.5	4.715×10^{-3}	17.5	3.951
12.5	5.235×10^{-3}	12.5	5.845×10^{-3}	12.5	3.572
7.5	4.178	7.5	9.741×10^{-3}	7.5	1.082×10^{-2}
5 h and 80 °C		5 h and 100 °C		5 h and 120 °C	
17.5	2.857	17.5	4.715×10^{-3}	17.5	3.951
12.5	5.23×10^{-3}	12.5	5.845×10^{-3}	12.5	3.572
7.5	4.178	7.5	9.741×10^{-3}	7.5	1.082×10^{-2}

Table 4: Volumetric mass transfer coefficients at extraction time 1, 3 and 5 h: temperature 80, 100 and 120 °C.

P (MPa)	$K \times 10^{-2}$ (s ⁻¹)	P (MPa)	$K \times 10^{-2}$ (s ⁻¹)	P (MPa)	$K \times 10^{-2}$ (s ⁻¹)
1 h and 80 °C		1 h and 100 °C		1 h and 120 °C	
17.5	0.291	17.5	1.741	17.5	0.394
12.5	0.596	12.5	2.049	12.5	0.586
7.5	1.218	7.5	2.486	7.5	1.037
3 h and 80 °C		3 h and 100 °C		3 h and 120 °C	
17.5	0.088	17.5	0.604	17.5	0.120
12.5	0.190	12.5	0.815	12.5	0.162
7.5	0.378	7.5	1.393	7.5	0.200
5 h and 80 °C		5 h and 100 °C		5 h and 120 °C	
17.5	0.046	17.5	0.238	17.5	0.046
12.5	0.089	12.5	0.336	12.5	0.082
7.5	0.222	7.5	0.571	7.5	0.166

the process parameters, exhibiting low standard deviation (0.29), high R-Squared (0.8405) and low PRESS (4.850) values. The quadratic response is expressed as follows:

$$K = \frac{1}{3600} \left[\begin{array}{l} -493.704 - 0.0848P + 12.919T - \\ 30.377t + (1.328 \times 10^{-5})P^2 - 0.066T^2 + \\ 2.576t^2 + (1.531 \times 10^{-4})PT + 0.0057Pt + \\ (2.419 \times 10^{-3})Tt \end{array} \right] \quad (11)$$

Where P, T, t and K are pressure [MPa], temperature [°C], extraction time [h] and volumetric mass transfer coefficient ($K = k_f a$) [s⁻¹].

The results related to statistical analysis and the mass transfer coefficients, K, are listed in Table 5.

The mass transfer coefficient versus the extraction pressure, extraction temperature and time of extraction is shown in Figure 1a-c. The extraction pressure, extraction temperature and time of extraction has a plateau at the

Table 5: Experimental program and results for the volumetric mass transfer coefficients of β -carotene supercritical extraction by CO₂ from CPO.

Run no.	P (MPa)	T (°C)	t (h)	$K \times 10^{-4}$ (s ⁻¹)
1	12.5	80	3	0.190
2	17.5	120	1	0.394
3	12.5	100	3	0.815
4	12.5	100	3	0.820
5	12.5	100	5	0.336
6	12.5	100	3	0.340
7	7.5	80	5	0.322
8	17.5	120	5	0.046
9	7.5	120	1	1.037
10	12.5	100	1	2.049
11	12.5	100	3	0.815
12	12.5	100	3	0.820
13	7.5	120	5	0.166
14	7.5	100	3	1.393
15	12.5	100	3	0.815
16	17.5	80	5	0.046
17	7.5	80	1	1.218
18	17.5	100	3	0.604
19	12.5	120	3	0.162
20	17.5	80	1	0.291

middle values of 12.5 MPa, 100 °C and 3 hour as shown in Figure 1a-c respectively. The significant of the investigated factor and their interaction were then examined. It is clearly seen that there is a high dependence of volumetric mass transfer coefficient on time of extraction as the absolute value of t (ratio of difference of sample mean to standard error of difference of sample means) is greater than 2.5 at P-level (probability of acceptance) less than 0.05 corresponding to variable time of extraction [15]. In a similar fashion, pressure was shown to be significant whereas temperature was shown to be not significant within the range studied. According to the second order terms of the three process parameters (P^2 , T^2 and t^2), T^2 only showed

a significant effect on volumetric mass transfer coefficient. According to the variables combination (PT, Pt and Tt), this model showed that these parameters had no significant effect on volumetric mass transfer coefficient.

Optimal processing conditions

The optimal processing conditions (P, T and t) for the volumetric mass transfer coefficients of β -carotene extraction from CPO were determined with a multiple regression relationship. The response surfaces have been shown in the figures (Figure 1 a-c). Figure 1a shows the volumetric mass transfer coefficient, K , as related to extraction pressure and extraction temperature,

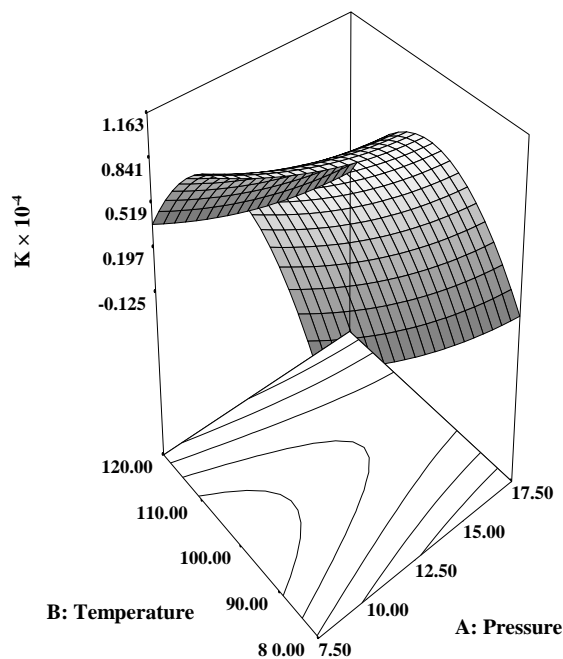


Fig 1a: Volumetric mass transfer coefficient as related to extraction temperature and extraction pressure (extraction time was set at 3 h)

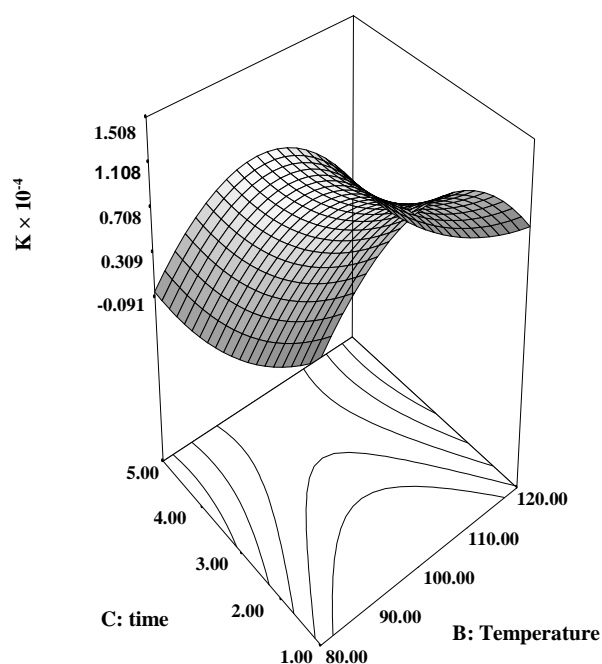


Fig. 1c: Volumetric mass transfer coefficient as related to extraction time and extraction temperature (extraction pressure was set at 12.5 MPa).

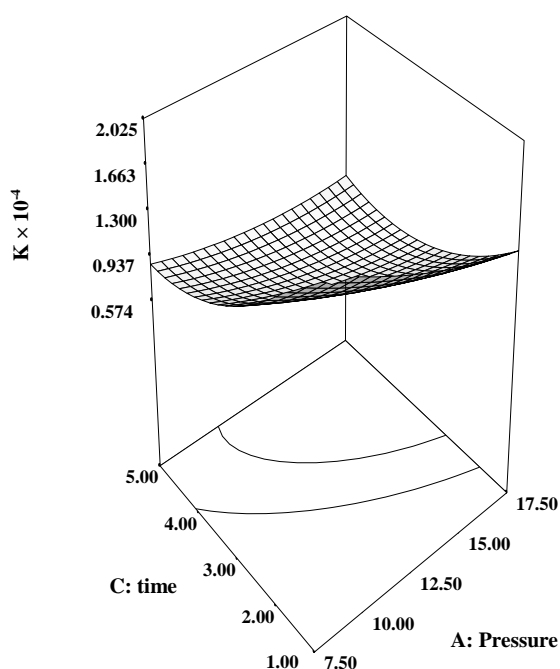


Fig. 1b: Volumetric mass transfer coefficient as related to extraction time and extraction pressure (extraction temperature was set at 100 °C).

with extraction time was fixed at 3 h. This figure showed that the maximum K of β -carotene extraction ($1.626 \times 10^{-4} \text{ s}^{-1}$) was obtained at pressure 7.5 MPa, temperature 100 °C and extraction time of 3 h (as actual factor). Fig. 1b shows the volumetric mass transfer coefficient as related to extraction pressure and extraction time (extraction temperature was set at 100 °C). This figure showed that the maximum K ($2.025 \times 10^{-4} \text{ s}^{-1}$) was obtained at extraction time of 1 h, pressure 7.5 MPa and temperature 100 °C (as actual factor). Figure 1c shows the volumetric mass transfer coefficient as related to extraction time and extraction temperature (extraction pressure was set at 12.5 MPa). This figure showed that the maximum K ($1.508 \times 10^{-4} \text{ s}^{-1}$) was obtained at extraction time 1h, extraction temperature 100 °C and extraction pressure 12.5 MPa (as actual factor).

To determine the optimal processing conditions of extraction of β -carotene from CPO, i.e., to determine the optimal values of P, T and t, the first partial derivatives of the regression equation was carried out with respect to P, T and t and set to zero. The point thus obtained is known as the stationary point, (P = 17.7 MPa, T = 100.5 °C and t = 3.9 h).

CONCLUSIONS

In this study, volumetric mass transfer factors such as diffusivity coefficients and mass transfer coefficients in supercritical fluid extraction of β -carotene from crude palm oil using carbon dioxide solvent in a bubbler extractor were determined. It was concluded that diffusivity coefficient in supercritical fluid extraction process was independent of extraction time but dependent on pressure and temperature. For the volumetric mass transfer coefficient, the highest value for β -carotene was experimentally obtained at $2.486 \times 10^{-2} \text{ s}^{-1}$ at the following conditions; pressure of 7.5 MPa, temperature of 100 °C and extraction time of 1 hour while based on statistical analysis, the optimum volumetric mass transfer coefficient value is at $6.700 \times 10^{-3} \text{ s}^{-1}$ at pressure of 17.7 MPa, temperature of 100.5 °C and extraction time of 3.9 h.

Nomenclature

D_{11}	Self diffusion of solvent, cm^2/s
D_c	Self diffusion of solvent at the critical point, cm^2/s
T_r	Reduced temperature, °C
ρ_r	Reduced density, g/cm^3
D_{12}	Binary diffusion, cm^2/s
V_{c1}	Critical volume of component 1, cm^3
V_{c2}	Critical volume of component 2, cm^3
M_1	Molecular weight component 1, g/mol
M_2	Molecular weight component 2, g/mol
M	Molecular weight, g/mol
T_c	Critical temperature, °C
P_c	Critical pressure, MPa
\dot{V}	Volumetric flow rate, cm^3/s
dC	Increase in solute concentration in the solvent stream, mol/cm^3
C	Concentration of solute in the solvent, mol/cm^3
C^*	Equilibrium concentration, mol/cm^3
k_f	Mass transfer coefficient, $\text{s}^{-1} \cdot (\text{cm}^2)^{-1}$
a	Mass transfer area, cm^2
τ	Extractor residence time, s
C_T	Concentration profile, mol/cm^3
K	Volumetric mass transfer coefficient, s^{-1}
T	Operating temperature, °C
P	Operating pressure, MPa
t	Extraction time, s
D_{AB}	Diffusivity coefficient of A (solvent) in B (solute), m^2/s

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