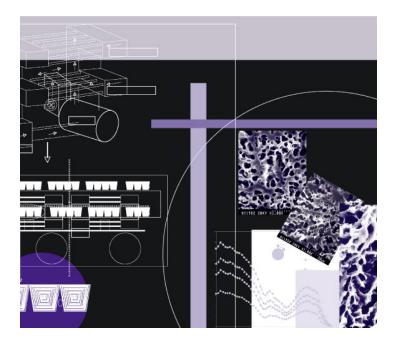
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Volume 85, Issue 3

ISSN 0260-8774

# journal of food engineering



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Journal of Food Engineering 85 (2008) 418-425

JOURNAL OF FOOD ENGINEERING

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# Mathematical modeling of vegetable oil—solvent extraction in a multistage horizontal extractor

M.E. Carrín\*, G.H. Crapiste

PLAPIQUI (Universidad Nacional del Sur – CONICET), Camino La Carrindanga Km 7, 8000 Bahía Blanca, Argentina

Received 11 December 2006; received in revised form 28 June 2007; accepted 5 August 2007 Available online 10 August 2007

#### Abstract

A new mathematical model of vegetable oil extraction in an industrial De Smet type extractor is proposed. Solvent extraction from a mobile bed of oilseed material was modeled considering different availability of oil in pellets, countercurrent cross-flow of the porous solid and the miscella, diffusion in the entire extraction field, mass transfer between pellets and miscella, miscella transport between the percolation sections, loading and drainage zones, and transient operational regime of the extractor. The mathematical model was solved numerically to predict oil concentration in miscella and pellets through the percolation sections and at the outlets. Model predictions were compared with experimental industrial data for sunflower oil extraction with hexane. Differentiation in two oil categories in terms of availability and extraction kinetics allows to represent the extraction phenomena in a better way than considering a single process. Consideration of dispersion fluxes improves the results without increasing the computing time. Saturation is reached in the stages near to loading zone when low miscella velocity is used.

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Keywords: Mathematical model; Mobile bed; Oilseeds; Percolation; Solvent extraction

# 1. Introduction

Solvent extraction is the primary means of extracting vegetable oil from oleaginous materials. Pretreated oilseeds (porous solid matrix) are put in contact with pure solvent (usually commercial hexane) or a solvent/oil mixture (miscella) for the oil to be transferred from the solid matrix to the fluid medium. Most industrial extractors available for solvent extraction of vegetable oil are countercurrent, multistage designs. Particularly, in the De Smet type extractor the solid particles are placed on a perforated base forming a bed that moves horizontally throughout the unit while the miscella flows through the bed (Kemper, 1997).

Although the principle of solvent extraction is relatively simple, it entails a complex mechanism, especially for oil-

bearing materials, because of the cellular structure of vegetable bodies. Therefore, it is difficult to explain all phenomena that take place during solvent extraction with a single theory (Rataus, 1997; Şaşmaz, 1996). Mathematical models with simplified equations have been proposed by several authors (Karnofsky, 1986; Majumdar, Samanta, & Sengupta, 1995; Pramparo, Gregory, & Mattea, 2002). Recently, a mathematical model for a countercurrent cross-flow system was presented, including loading and drainage zones (Veloso, Krioukov, & Vielmo, 2005). However, it does not consider changes in oil flux resistances along the extraction process, neither the mass transfer resistance in the solid matrix. Further more, it assumes equilibrium state between pore and solid phases in any point of the field, including loading zone.

The objectives of the present study were: (i) to represent the extraction process by a two-dimensional transient model introducing the concept of different oil categories, and (ii) to analyze the effects of some parameters as

<sup>\*</sup> Corresponding author. Tel.: +54 291 4861700; fax: +54 291 4861600. E-mail address: mcarrin@plapiqui.edu.ar (M.E. Carrín).

Nomenclature						
$a_{\rm p}$	pellet surface area per volume unit (m <sup>-1</sup> )	X	bed width (m)			
$\stackrel{\scriptscriptstyle{\mathrm{P}}}{C}$	oil concentration in bulk phase (oil kg/miscella	X	oil molar fraction in miscella			
	kg)	$y^*$	dimensionless horizontal coordinate $(=y/W)$			
$C_{\rm s}$	oil concentration in solid-occluded miscella	z*	dimensionless vertical coordinate $(=z/L)$			
	phase (oil kg/inert solid kg)					
d	diameter (m)	Greek	t letters			
D	oil diffusivity (m <sup>2</sup> /s)	3	porosity			
$D_z$	dispersion coefficient (m <sup>2</sup> /s)	$\rho$	bulk phase density (kg/m <sup>3</sup> )			
$d_{\mathrm{p}}$	pellet equivalent diameter (m)	$ ho_{ m s}$	solid density (kg/m <sup>3</sup> )			
$dz^*$	dimensionless control volume height	$\mu$	bulk phase viscosity (Pa s)			
K	equilibrium constant $(=C/C_s)$					
$k_{ m L}$	convective mass transfer coefficient (m/s)	Subscripts				
L	bed height (m)	0	initial state			
M	molecular weight	1	first stage			
$m_{\rm s}$	solid mass flow rate (kg/s)	b	bed			
n	oil diffusive flux from pellet to bulk phase (kg/	cr	critic oil			
	$s m^2$ )	d	drainage stage			
Pe	Peclet number $(=v_{\rm m}d_{\rm p}/D_z)$	fd	first drainage point of the stage			
PN	pump number or number of trays in the extrac-	h	hexane			
	tor	i	oil category			
$t^*$	dimensionless time ( $=v_{\rm m}t/L$ )	ip	internal particle			
$t_{ m r}^*$	dimensionless residence time to each section of	L	bottom bed			
	the bed	ld	last drainage point of the stage			
и	velocity of the transporter (m)	m	miscella in trays			
V	molar volume (m³/mol)	O	oil			
$v_{ m m}$	vertical bulk phase velocity in the bed (m/s)	p	pellet			
$V_{\rm t}$	tray volume (m <sup>3</sup> )	PN	pump number or number of trays in the extrac-			
$v_y^*$	dimensionless horizontal bulk phase velocity		tor			
	$(=v_y/v_m)$	po	pellet pore			
$v_z^*$	dimensionless vertical bulk phase velocity (= $v_z$ /	S	stage			
	$v_{ m m}$ )	$\propto$	infinite time			
W	bed length (m)					
w	oil volumetric fraction in miscella	Superscripts				
$W_{\rm d}$	drainage length (m)	a	average at a fixed horizontal position of the bed			
$W_{\rm e}$	stage length (m)	0	initial state			

miscella velocity and oil concentration of different categories on the behavior of the process. Some results predicted by the model were compared with experimental data.

#### 2. Extraction model

In a De Smet type extractor, two media are involved: (a) a porous media of pretreated (mechanically crushed and pelletized) oleaginous material with two porosity types (bed porosity and pellet porosity) that moves horizontally on a perforated band; and (b) an oil–solvent miscella that moves in countercurrent cross-flow.

Fig. 1 presents a schematic drawing of the extractor. The porous solid enters in the extractor through tube 1 in the loading zone, fills the bed to a certain height and moves horizontally through the extractor until reaching the drainage zone (6) where it leaves the extractor. The liquid phase

enters as solvent in the extractor through tube 2 spraying the extraction stage nearest to the drainage zone (I), moves through percolation sections, trays and pumps (from I to N) as miscella of increasing oil concentration, and leaves the extraction unit as oil enriched miscella through the discharge tube 3. Each percolation section or stage and loading zone are determined by the quality of sprayed miscella.

In the loading zone (5), the concentrated miscella fills out the spaces between pellets and permeates the pore phase. In the drainage stage (6), the miscella flows from the spaces between pellets, but still remains in the pore phase.

In all the extraction process, part of the solvent that flows through the bed goes into the pellets through their pores, dissolves the oil and returns to the bulk phase from where it drains from the bed. The oil in the pellet diffuses toward the pellet surface entering the bulk phase by a convective mechanism produced by a concentration difference

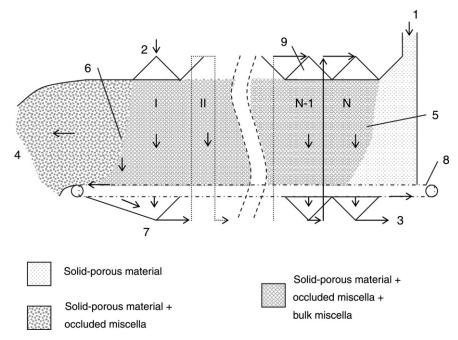


Fig. 1. Scheme of the process in the extractor: (1) porous media inlet; (2) solvent inlet; (3) miscella exit; (4) porous media exit; (5) loading zone; (6) drainage zone; (7) tray; (8) transporter; (9) miscella distributor; (I) to (N) extraction stages (tray numeration is the same as extraction stage numeration).

between the pellet surface and the moving bulk phase. Then, convective movement to the bottom of the bed carries the oil away. In the bulk phase, an oil concentration gradient develops by the increasing concentration of oil that the miscella obtains as it flows down the bed. This concentration gradient induces a diffusive mass transfer of solute in the direction from higher to lower solute concentration, producing a dispersive flow of oil from the bottom toward the top of the bed which is in the opposite direction to the convective flow of the miscella (Majumdar et al., 1995). In each section, the oil concentration in both bulk and solid-pore phases are not uniform in the space during the extraction.

The aim of the model presented in this work is to predict the oil concentration profiles in miscella and solid in the extraction unit including loading and drainage zones, the oil concentration in miscella pumped in each stage, and the average oil concentration in solid leaving the drainage zone, from the startup of the extraction process until the stationary state.

The assumptions made in developing the proposed model were:

- the temperature was constant throughout the process;
- vegetable oil was regarded as a single component because all glycerides are highly soluble in hexane, but with different mass transfer properties in solid structure of particles depending on their accessibility (Fan, Morris, & Wakeham, 1948);
- the extraction unit was considered as a solid bed made of isotropic porous particles of equivalent diameter  $(d_p)$ , with bed porosity  $(\varepsilon_b)$  and pellet porosity  $(\varepsilon_p)$  constants and uniforms through the bed;

- the extraction was considered as a mass transfer process between the solid (diffusion) and the bulk fluid phase (convection);
- oil volume extracted from solid material was replaced by the same volume of solvent;
- the horizontal velocity of the porous media was equal to the velocity of the transporter, *u*;
- the upper surface of a extraction stage was covered by miscella that leaves from the pump of the close tray with lower oil concentration, excepting the first stage where solvent/miscella was pumped from an external tank;
- the bulk phase miscella velocities are constant while the oil concentrations can be modified as time-dependent;
- the vertical velocity of percolation, v<sub>m</sub>, occurs under gravity and is determined by the properties of both the miscella and the porous media, and is considered constant in each section;
- the miscella drag velocity of the bulk phase in the horizontal direction is considered as constant in the entire extraction field and equal to the solid velocity. This velocity creates a horizontal flow of bulk phase in the direction of bed movement.
- *Drainage zone*. Only liquid bulk phase is drained and this miscella is collected in the same tray than the miscella from the first extraction section. Oil transfer between pore and bulk phases occurs while there is bulk phase surrounding the pellets.
- Loading zone. The solid-porous material enters in this zone with known mass flow rate and mass concentration and without any solvent. As miscella is sprayed, the bed porous are occupied by miscella and oil transfer between pellets and miscella begins. At the bottom of

this section, a part of the solid-porous material is without miscella because of the bed movement and residence time. So, the collected miscella flow is lower than that sprayed over in this section.

# 2.1. Oil transfer equation

Pellets are formed by joined solid particles in porous matrixes. Oil in solid particles is thought to be composed by two categories (Fan et al., 1948): (C1) free oil that is easily extracted from the solid particles, and (C2) bounded oil that is hardly extracted. Maximum concentration of C2-oil in particle is called critic oil content,  $C_{\rm scr}$ . Oil is extracted from the porous solid by parallel diffusion of both oil categories into particles and series diffusion of these with flux in matrix porous. Applying mass balance to solid-porous particles at stationary state when convective mass transport occurs between the matrix surface and the bulk phase, the i-category oil diffusive flux leaving porous matrix can be expressed as (Wakao & Kaguei, 1982):

$$n_{\rm i} = \frac{d_{\rm p}}{6} \frac{\rho}{\left(\frac{d_{\rm p}}{6k_{\rm L}} + \frac{1}{K_{\rm x}E_{\rm i}}\right)} (KC_{\rm si} - C_{\rm i}) \tag{1}$$

with  $E_{\rm f}=\frac{3}{\phi}(\coth\phi-1/\phi),~\phi=\frac{d_{\rm p}}{2}(\frac{K_{\rm x}}{D_{\rm oh}})^{0.5},~K_{\rm x}=\frac{D_{\rm f}\xi}{\rho K d_{\rm ip}},~{\rm and}~\xi=\frac{4\rho_{\rm s}}{d_{\rm po}c_{\rm p}}$  where  $E_{\rm f}$  is the diffusion effectiveness factor and  $\phi$  is the Jüttner modulus (which relates particle  $(D_{\rm i})$  and liquid  $(D_{\rm oh})$  diffusion).  $E_{\rm f}$  reaches the value of 1 when the process is controlled by solid internal diffusion and the value of  $3/\phi$  when liquid diffusion is the controlling process.

#### 2.2. Mass balances

The mathematical model includes the extraction, drainage and loading zones and the trays. Oil mass balances in bulk miscella and solid-porous material (including occluded miscella) in a bed portion led to the following equations expressed in a dimensionless form:

# • Bulk phase

$$\begin{split} \frac{\partial(\rho C_{\rm i})}{\partial t^*} &= -\frac{\partial(v_z^* C_{\rm i} \rho)}{\partial z^*} - \left(\frac{L}{W}\right) \frac{\partial(v_y^* C_{\rm i} \rho)}{\partial y^*} + \left(\frac{d_{\rm p}}{L}\right) \\ &\times \left(\frac{\partial}{\partial z^*} \left(\frac{v_z^* \rho}{Pe} \frac{\partial C_{\rm i}}{\partial z^*}\right)\right) + \left(\frac{d_{\rm p}}{L}\right) \left(\frac{L}{W}\right)^2 \\ &\times \left(\frac{\partial}{\partial v^*} \left(\frac{v_y^* \rho}{Pe} \frac{\partial C_{\rm i}}{\partial v^*}\right)\right) + \left(\frac{1 - \varepsilon_{\rm b}}{\varepsilon_{\rm b}}\right) \left(a_{\rm p} \frac{L}{v_{\rm m}}\right) n_{\rm i}. \end{split} \tag{2}$$

The terms on the right-hand side of Eq. (1) represent the movement in the vertical and horizontal directions, dispersion in the vertical and horizontal directions, and mass transfer between bulk and solid-porous phases, respectively.

• Solid-porous (pellet) and occluded miscella

$$\frac{\partial C_{\rm si}}{\partial t^*} = -\frac{a_{\rm p}}{\rho_{\rm s}} \frac{L}{v_{\rm m}} n_{\rm i} - \frac{m_{\rm s}}{\rho_{\rm s} dz^* XWL (1 - \varepsilon_{\rm b})} \left(\frac{L}{v_{\rm m}}\right) \frac{\partial (C_{\rm si})}{\partial y^*},\tag{3}$$

where the first term on the right-hand reflects the mass transfer between the bulk and pellet phases, and the second term corresponds to the movement in the horizontal direction.

#### • Trays

The miscella flow pours into a tray with non-uniform icategory oil concentration distribution,  $C_{\rm iL}$ . Considering a rapid mixture of the miscella inside the tray, it can be assumed that the concentration in the tray is uniform, but time-dependent for the transient regime. Applying oil conservation law to the tray volume  $V_{\rm t}$ , the equation to account for changes in the miscella for i-category oil concentration in the intermediate trays ( $C_{\rm mis}$ ) is

$$\frac{\mathrm{d}(C_{\mathrm{mis}}\rho_{\mathrm{ms}})}{\mathrm{d}t^{*}} = (\varepsilon_{\mathrm{b}}LX/V_{t}) \left(W \int_{y_{\mathrm{fds}}^{*}}^{y_{\mathrm{ids}}^{*}} (C_{\mathrm{iL}}\rho_{\mathrm{L}}) \mathrm{d}y^{*} - W_{\mathrm{e}}C_{\mathrm{mis}}\rho_{\mathrm{ms}}\right). \tag{4}$$

The first tray (1) receives not only the miscella that flows out from the first stage, but also the drainage miscella. So, the corresponded equation to account for changes in the miscella for i-category oil concentration in the first tray ( $C_{\rm mil}$ ) is

$$\frac{d(C_{\min}\rho_{m1})}{dt^*} = (\varepsilon_b LX/V_t) \left( W \int_{y_{\text{fdl}}^*}^{y_{\text{idd}}^*} (C_{\text{iL}}\rho_{\text{L}}) dy^* - (W_e + W_d) C_{\min}\rho_{\text{m1}} \right). \tag{5}$$

For the last tray (PN), the outlet miscella mass concentration is not established by the accumulation in a tray but by the integration over the drainage zone of this tray. The following expression results for i-category oil concentration in this miscella ( $C_{\rm miPN}$ ):

$$C_{\text{miPN}}\rho_{\text{mPN}} = \int_{y_{\text{fdPN}}^*}^{y_{\text{idPN}}^*} (C_{\text{iL}}\rho_{\text{L}}) dy^* / \int_{y_{\text{fdPN}}^*}^{y_{\text{idPN}}^*} dy^*.$$
 (6)

Average oil concentration in solid-occluded miscella phase at a fixed horizontal position of the bed  $(C_{si}^a)$  is evaluated through:

$$C_{\rm si}^{\rm a} = \int_0^1 C_{\rm si} \mathrm{d}z^* \tag{7}$$

# 2.3. Physical properties and transport coefficients

Convective mass transfer coefficient,  $k_{\rm L}$ , was obtained from dimensionless correlations in terms of the Sherwood number,  $Sh = k_{\rm L}d_{\rm p}/D_{\rm ho}$ , by (Treybal, 1981)

$$Sh = 2.4\varepsilon_b Re^{0.34} Sc^{0.42}$$
 for  $0.08 \prec Re \prec 125$   
 $Sh = 0.442\varepsilon_b Re^{0.69} Sc^{0.42}$  for  $125 \prec Re \prec 5000$ . (8)

The Reynolds, *Re*, and Schmidt, *Sc*, numbers were upgraded by means of the following equations

$$Re = \frac{\rho v d_{\rm p}}{\mu} = Re^0 \left(\frac{\rho}{\rho^0}\right) \left(\frac{\mu^0}{\mu}\right),\tag{9}$$

$$Sc = \frac{\mu}{\rho D_{\text{oh}}} = Sc^0 \left(\frac{D_{\text{oh}}^0}{D_{\text{oh}}}\right) \left(\frac{\rho^0}{\rho}\right) \left(\frac{\mu}{\mu^0}\right),\tag{10}$$

where  $\mu$ : bulk phase viscosity, subscripts h and o refer to hexane and oil, respectively, and superscript 0 indicates the initial state.

The Peclet number was obtained by (Butt, 1980)

$$\varepsilon_{\rm b} Pe = 0.2 + 0.011 (\varepsilon_{\rm b} Re)^{0.48}.$$
 (11)

Physical and transport properties of miscella as function of oil concentration were evaluated by (Perry & Green, 1984; Rice & Hamm, 2002)

$$\rho = \frac{M_{\rm h} M_{\rm o}}{V_{\rm h} M_{\rm o} + (V_{\rm o} M_{\rm h} - V_{\rm h} M_{\rm o}) C} \tag{12}$$

$$\mu = \mu_h^{1-w} \mu_0^w \tag{13}$$

$$\frac{D_{\text{oh}}}{D_{\text{oh}}^{0}} = \left[ \frac{(V_{\text{h}}^{2/3} + (3V_{\text{o}})^{2/3})\mu_{\text{h}}V_{\text{o}}}{(V_{\text{o}}^{2/3} + (3V_{\text{h}})^{2/3})\mu_{\text{o}}V_{\text{h}}} \right]^{x-x^{0}}.$$
(14)

 $D_1$ ,  $D_2$  and  $C_{\rm scr}$  were obtained experimentally through batch extraction of sunflower flour with hexane at 56 °C. Data of oil concentration in particles as a function of time were used to determine the parameters by comparing the solution of the unsteady-state diffusion equation (Crank, 1967; Şaşmaz, 1996) and modeling the diffusion process as a parallel extraction one with the resulting equation:

$$\frac{C_{s0} - C_{s}}{C_{s0} - C_{s\infty}} = 1 - \left(1 - \frac{C_{scr}}{C_{s0} - C_{s\infty}}\right) \exp\left(-\frac{4D_{1}\pi^{2}t}{d_{ip}^{2}}\right) - \left(\frac{C_{scr}}{C_{s0} - C_{s\infty}}\right) \exp\left(-\frac{4D_{2}\pi^{2}t}{d_{ip}^{2}}\right).$$

The equilibrium relationship between the oil concentration of miscella and the oil concentration in solid, K, was evaluated varying the solvent–solid relation at the same experimental conditions when stationary state was reached.

### 2.4. Initial and boundary conditions

As bed is not filled with solution at the beginning of the extraction process, initial conditions were not evaluated at time zero. This situation was represented doing initial conditions function of dimensionless residence time to each section of the bed,  $t_r^*$ , as follows:

$$\begin{split} t^* &< t_{\rm r}^*, \quad C_{\rm i}(y^*,z^*,t^*) = 0, \quad C_{\rm si}(y^*,z^*,t^*) = C_{\rm si}^0, \\ y^* &= 0,\dots,1, \quad z^* = 0,\dots,1 \\ t^* &= t_{\rm r}^*, \quad \frac{\partial C_{\rm i}(y^*,z^*,t_{\rm r}^*)}{\partial z^*} = 0, \quad C_{\rm si}(y^*,z^*,t_{\rm r}^*) = C_{\rm si}^0, \\ y^* &= 0,\dots,1, \quad z^* > 0 \\ C_{\rm i}(y^*,0,t_{\rm r}^*) = C_{\rm i}^0, \quad C_{\rm si}(y^*,z^*,t_{\rm r}^*) = C_{\rm si}^0, \quad y^* = 0,\dots,1. \end{split}$$

When the bed is filled with miscella, the boundary conditions for the bulk and solid-porous phases are:

• for right and left-hand of the extraction field

$$\begin{aligned} \frac{\partial C_{\mathbf{i}}(0,z^*,t^*)}{\partial y^*} &= 0 & \frac{\partial C_{\mathbf{i}}(1,z^*,t^*)}{\partial y^*} &= 0 & z^* &= 0,\dots,1 & t^* \geqslant 0 \\ C_{\mathbf{si}}(0,z^*,t^*) &= C_{\mathbf{si}}^0, & z^* &= 0,\dots,1 & t^* \geqslant 0 \end{aligned}$$

• for the upper and bottom boundaries of the field

$$\begin{split} t^* > t_{\rm r}^*, \quad C_{\rm i}(y^*,0,t^*) &= C_{\rm mi,s-1}(t^*), \quad y^* = 0,\dots,y_{\rm fd1}^* \\ C_{\rm i}(y^*,0,t^*) &= C_{\rm i}^0, \quad y^* = y_{\rm fd1}^*,\dots,y_{\rm ld1}^* \\ \frac{\partial C_{\rm i}(y^*,0,t^*)}{\partial z^*} &= 0, \quad y^* = y_{\rm ld1}^*,\dots,1 \\ \frac{\partial C_{\rm i}(y^*,1,t^*)}{\partial z^*} &= 0, \quad y^* = 0,\dots,1 \end{split}$$

where  $y_{\text{fd1}}^* = (1 - 1/\text{PN})(1 - W_d/W), y_{\text{ld1}}^* = (1 - W_d/W).$ 

# 2.5. Resolution procedure

The set of equations (1)–(14), with the initial and boundary conditions listed above, constitute the proposed model describing the behavior of DeSmet-type extractor. The model was solved numerically using the finite differences method (Davis, 1984) to discretize the spatial derivatives, and finite sums to replace integral terms. The obtained ordinary differential equations were solved through the fourth-order Runge–Kutta numerical method, with Mat-Lab 5.3.

#### 3. Materials and methods

All experimental determinations were made with sunflower pellets and miscella obtained from different sections of the industrial extractor that were kindly provided by OMHSA Company. Results are expressed as average value of four replicates  $\pm$  standard deviation.

The oil content of the solids was determined according to AOCS method Ac 3-44. The miscella concentration was determined by evaporating the solvent to constant weight.

The bed and pellet porosities were calculated from bed and pellet densities and pellet and particle (disaggregated pellet) densities, respectively, determined with a pyncnometer according to AOCS method Cc 10a-25 with modifications. When bed gravity was determined only pellets were introduced into pyncnometer of known volume. On the other hand, when pellet (particle) gravity was determined weighted pellets (particles) were covered by glycerin and then pellets (particles) volume was calculated as the difference between the volumes of pyncnometer and glycerin.

Pellet specific areas were calculated from diameter and length measures of pellets and equivalent diameter of a

sphere was calculated from the obtained average specific area.

Particle diameter was calculated adding the product of the weigh fraction obtained sifting the disaggregated pellet and the corresponding sieve diameter. Sieves of 840, 715, 505 and 358.5 µm were used.

#### 4. Results and discussion

Results of numerical solution from the proposed model are compared with experimental data of composition obtained from miscella and pellets collected at stationary state operation in a De Smet type sunflower oil industrial extractor with the following characteristics: W=14.5 m, L=1.8 m, X=2.4 m, PN = 8,  $W_{\rm d}=2.3 \text{ m}$ , u=0.003 m/s,  $v_{\rm m}=0.0125 \text{ m/s}$ ,  $V_{\rm t}=2.31 \text{ m}^3$ .

Properties of the porous media (sunflower pellets) were:  $\varepsilon_{\rm p} = 0.24 \pm 0.03, \quad \varepsilon_{\rm b} = 0.4 \pm 0.08, \quad d_{\rm p} = 0.015 \pm 0.002 \text{ m}, \quad C_{\rm so} = 0.2419 \pm 0.0037, \quad d_{\rm ip} = 6.18 \times 10^{-4} \text{ m}, \quad \rho_{\rm s} = 742.4 \pm 3.7 \text{ kg/m}^3, \quad K = 1.5 \pm 0.08, \quad C_{\rm scr} = 0.01, \quad D_1 = 9.674 \times 10^{-5} \text{ m}^2/\text{s}, \quad D_2 = 2.13 \times 10^{-10} \text{ m}^2/\text{s}. \quad \text{Diffusivity values of oil in}$ liquid phase (solvent) reported at the literature (Fan et al., 1948; Şaşmaz, 1996) have an order of  $10^{-8}$  $10^{-9}$  m<sup>2</sup>/s. It is clear that the  $D_1$ -value has not the diffusion physical sense. However, we used it in the proposed model as a characteristic parameter of the particle washing process that occur with this oil category. This assumed analogy between diffusion and washing process in particles let us have a unique model to describe the behavior of both oil categories without despise the diffusion of C1-oil between pore pellets. Simulations to determined the sensibility of the model to  $d_{po}$  value were done between  $2d_{ip}$  and  $10d_{ip}$ , finding that the result differences were little (maximum error = 2%). So, a  $d_{po}$  value of  $10 d_{\rm ip}$  was used.

Average oil concentrations in miscella trays and pellets leaving each extraction stage are shown in Table 1. Model outputs showed a good agreement with industrial measurements since a *t*-test analysis showed that the differences between experimental and simulated data

were not significant ( $\alpha = 0.05$ ). We can conclude that the mathematical model developed in this study closely represented the complex phenomena taking place during the extraction process. It can be observed that more than 85% of available oil is extracted at the last three stages of extraction while C2 oil is mainly extracted at

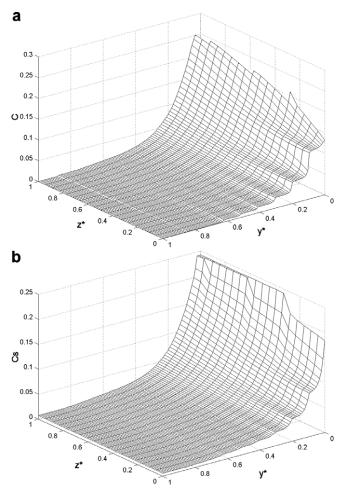


Fig. 2. Oil concentration profiles in miscella (a) and pellets (b) in the extractor for steady-state operation.

Table 1 Simulation results and experimental data of average oil concentration in miscella trays ( $C_{\rm m}$ ) and pellets ( $C_{\rm sal}$ )

Stage	Simulation model					Experimental			
	Without dispersion			With dispersion					
	$\overline{C_{ m m}}$	$C_{\mathrm{sa}}$	$C_{\mathrm{m}}$	$C_{\rm sal}^{\ \ a}$	$C_{\mathrm{sa2}}^{}a}$	$C_{\mathrm{sa}}$	$C_{\rm m}^{}$	$C_{\mathrm{sa}}^{}\mathrm{b}}$	
1	0.0012	0.0067	0.0012	0.00086	0.0057	0.00656	$0.0024 \pm 8 \times 10^{-4}$	$0.0079 \pm 9 \times 10^{-4}$	
2	0.0053	0.0091	0.0050	0.0016	0.0072	0.0088	$0.0049 \pm 7 \times 10^{-5}$	_	
3	0.0095	0.013	0.0088	0.0040	0.0085	0.0125	$0.0085 \pm 2 \times 10^{-4}$	_	
4	0.0157	0.0169	0.0142	0.0067	0.0095	0.0162	$0.0145 \pm 2 \times 10^{-4}$	_	
5	0.0288	0.0223	0.0258	0.0111	0.01	0.0211	$0.0250 \pm 6 \times 10^{-4}$	_	
6	0.0582	0.0333	0.0527	0.0210	0.01	0.0310	$0.0449 \pm 6 \times 10^{-3}$	_	
7	0.1242	0.0581	0.1145	0.0441	0.01	0.0541	$0.0993 \pm 0.01$	_	
8	0.2258	0.1146	0.2127	0.0980	0.01	0.1080	$0.2237 \pm 8 \times 10^{-3}$	_	

<sup>&</sup>lt;sup>a</sup> Oil category in particles: (1) free and (2) bounded oils.

<sup>&</sup>lt;sup>b</sup> Average values and standard deviations of four replicates.

Table 2 Effect of miscella velocity  $(v_m)$  on simulated average oil concentration in miscella trays  $(C_m)$  and pellets  $(C_{sa})$ 

Stage	$v_{\mathrm{m}}=0.0125~\mathrm{m/s}$				$v_{\rm m} = 0.0100 \; {\rm m/s}$			
	$\overline{C_{ m m}}$	$C_{\mathrm{sa1}}^{}\mathrm{a}}$	$C_{\mathrm{sa2}}{}^{\mathrm{a}}$	$C_{\mathrm{sa}}$	$C_{ m m}$	$C_{\mathrm{sa1}}{}^{\mathrm{a}}$	$C_{\mathrm{sa2}}{}^{\mathrm{a}}$	$C_{\mathrm{sa}}$
1	0.0012	0.00086	0.0057	0.00656	0.0059	0.0019	0.0082	0.0101
2	0.0050	0.0016	0.0072	0.0088	0.0387	0.0106	0.0098	0.0204
3	0.0088	0.0040	0.0085	0.0125	0.0725	0.0323	0.01	0.0423
4	0.0142	0.0067	0.0095	0.0162	0.1087	0.0553	0.01	0.0653
5	0.0258	0.0111	0.01	0.0211	0.1487	0.0803	0.01	0.0903
6	0.0527	0.0210	0.01	0.0310	0.1945	0.1081	0.01	0.1181
7	0.1145	0.0441	0.01	0.0541	0.2486	0.1404	0.01	0.1504
8	0.2127	0.0980	0.01	0.1080	0.2959	0.1792	0.01	0.1892

<sup>&</sup>lt;sup>a</sup> Oil category in particles: (1) free and (2) bounded oils.

the first four stages, when miscella with low oil concentration is used. Results of simulation obtained considering dispersion terms were more closed to experimental data than those obtained without considering dispersion and the computing time was not modified. The bigger difference was obtained at stage 7 where 1.5% of difference was calculated with dispersion flux instead of 2.5% of difference obtained without these terms. So, that simplification is not a recommended practice for a rigurous modeling. The mean accuracy of the results predicted with the proposed model is 0.46%, varying from 0.01% to 1.52%. Veloso et al. (2005) modeled solvent extraction of soy oil considering equilibrium between the pore and solid phases in any point of the extraction and found a mean accuracy of 1.61% varying from 0.1% to 4.8%.

The evolution of oil concentrations in miscella and pellets throughout the extractor showed a wave behavior at the top of the bed  $(z^*=0)$  during the transient state because of the change in miscella quality between two stages. This behavior disappears at the bottom of the bed  $(z^*=1)$ , where a monotonous diminution of oil concentrations is observed. Wave magnitudes are bigger when processing time is smaller (data no shown). At steady-state, this behavior is observed only in stages nearest the loading zone where extraction of free oil (C1 category) occurs (Fig. 2). From middle bed to drainage zone  $(y^*=0.5-1)$ , a plateau is obtained because of the slow transfer of the bounded oil (C1 category).

The influence of miscella velocity  $(v_{\rm m})$  is seen in Table 2. It shows that oil concentration in both pellets and miscella decreased as  $v_{\rm m}$  increased. A higher  $v_{\rm m}$  implies not only a higher convective coefficient that increases mass transfer, but also a faster replacement of more concentrated miscella by less one through the bed. For lower  $v_{\rm m}$ , the oil released from the solids to the stagnant occluded miscella in pores is not easily convected away by the bulk phase. This effect does not allow the extraction of the C2 category oil because of the miscella saturation, except at two first stages. It can be observed that less than 50% of available oil is extracted at the last three stages of extraction, compared with more than 85% of extraction reached to higher  $v_{\rm m}$ .

Differentiation of oil content in two categories improves simulation results in stages of less concentrated miscella. Average oil concentration in pellet in the extraction bed at the outlet was 0.16% without that differentiation instead of the 0.66% obtained when differentiation was used (experimental value is 0.79%).

Hard extraction or bounded oil category has the control of the process at the end of the extraction equipment. Pretreatment of solid material could modify the relation between both oil categories. Simulation results for pellets with higher  $C_{\rm scr}$  (0.03 instead of 0.01) showed a less effective process, with a higher average oil concentration in the meal (1.67% instead of 0.66%) and lower oil concentration in the outlet miscella (20.13% instead of 21.27%).

In conclusion, a two-dimensional nonsteady-state mathematical model for vegetable oil-solvent extraction in a De Smet type extractor was developed. The model can be used to predict oil concentration profiles in miscella and pellets throughout the extractor and in the outlets under different operating conditions, permitting knowing the expected separation in an industrial processing without the experimentation. Use of the model can be extended, with little modifications, to study the differential extraction of some oil minor components of industrial interest, as waxes and phospolipids, provided their extraction kinetic are known. Generalizing the concepts to solid-liquid extraction, the analogy introduced to treat the wash-period as a diffusion process could be used to simulate non-ideal separations and to obtain the solute critical content responsible of that non-ideal behavior.

#### Acknowledgements

The authors would like to thank the Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), the Agencia Nacional de Promoción Científica y Tecnológica (FONCyT) and the Universidad Nacional del Sur (UNS) of Argentina for the financial assistance to this work. The authors also acknowledge the OMHSA Company for possibility to obtain experimental data in an industrial extractor.

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