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Contributions to supercritical extraction of vegetable substrates in Latin America

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Abstract

This manuscript summarizes basic and applied research on phase equilibrium and mass transfer kinetics involved in high-pressure CO_2 extraction of solid substrates. Most examples relate to the extraction of lipids and essential oils from native Latin American plants. Extraction rates of vegetable matrices depend on the external mass transfer coefficient (k_f), effective solute diffusivity in the solid substrate (D_e), solute solubility in high-pressure CO_2, and solute binding to the solid matrix. The initial stages of the extraction process depend on an operational solubility that is close to the thermodynamic solubility (c_{sat}) in the case of lipid extraction from oil-containing plant material, but lower than c_{sat} in the case of essential oils, due probably to stronger interactions between essential oils than lipids and the solid matrix. Experimental values of k_f exhibited considerable scattering and were several orders of magnitude smaller than corresponding values from literature correlations for the dissolution of solids or evaporation of liquids from films with supercritical fluids (SCFs), due to underestimation of the contribution of internal (solid phase) mechanisms to the total resistance to mass transfer and other aspects. D_e values were 10–10^3 or 10^2–10^5 times smaller than binary diffusion coefficients of lipids and essential oils, respectively, in high-pressure CO_2, suggesting very pronounced limitations to mass transfer within the solid matrices in both cases. The integration of this information for the modeling, simulation, and scaling-up of laboratory data is thoroughly discussed. Finally, an example of economic feasibility is given for the installation of a SCF extraction plant for the recovery of lipids from wheat germ.

Keywords: Effective diffusivity; External mass transfer coefficient; Essential oils; Herbs; Latin America; Lipids; Modeling; Oilseeds; Solubility; Extraction; Spices; Supercritical CO_2; Supercritical extraction

1. Introduction

del Valle and Aguilera (1999) reviewed the use of high-pressure carbon dioxide (CO_2) for the selective extraction of essential oils, pungent principles, carotenoid pigments, antioxidants, antimicrobials, and related substances from spices, herbs, and other plant materials. Discussion was focused on the potential application of supercritical fluid (SCF) extraction (SCFE) for the production of value-added ingredients for the food, pharmaceutical, and perfumery industries in Latin America. By 1998, SCFE had experienced a very pronounced development over a two-decade span, which had resulted on many industrial applications mostly in Europe and North America, but none in Latin America, where there were just incipient efforts of basic and applied research. Over the past 5 years, many other applications have arisen which have not been limited to developed countries, e.g., concerted research efforts in East Asia have

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Nomenclature

a_p	specific surface area of a solid substrate particle
A	parameter in Eq. (4)
c_f	concentration of solute in the SCF
c_f^*	concentration of solute in a SCF film that is in equilibrium with a solid surface
c_k	critical solute concentration in the solid for which the proportionality in the partition of the solute between the two phases is lost
c_m	monolayer concentration of solute on the solid
c_s	concentration of solute in the solid or adsorbed onto the solid
c_{sat}	concentration of the solute in a saturated SCF phase
c_{so}	initial concentration of solute adsorbed onto the solid
$\bar{c}_s$	average solute concentration in the solid matrix
C	capacity of an SCFE plant
d	connectedness of a network of pores
d_c	cell diameter
d_E	diameter of the extraction vessel
d_p	diameter of a particle
d_{pore}	diameter of the pore in a particle
D_{12}	is the binary diffusion coefficient of the solute in the solvent
D_m	heterogeneity of the porous space
D_e	effective diffusion coefficient in a solid substrate
D'_e	diffusivity of a solute in a vegetable matrix
D_L	axial dispersion coefficient of solute in the SCF
f	function relating the composition of a solid substrate (c_s) with that of a SCF phase with which it is equilibrated
f_T	temperature factor correction for the solubility
F_e	correction factor in Eq. (5)
F_λ	restriction factor for solute–matrix interaction
I	purchasing cost of a SCFE plant
k	scaling exponent factor (Eq. (8))
k_i	internal mass transfer coefficient
k_f	film mass transfer coefficient
K_L	equilibrium sorption coefficient (Eq. (1))
L	length of the packed bed.
MW	molecular weight
n	exponent of Eq. (4)
p_{cm}	critical pressure of the mixture
p_{nom}	nominal pressure
PCF	pressure-correction factor
q	specific mass flow of SCF
Q	maximal pumping flow of a SCFE plant
R_{ext}	external resistance to mass transfer
R_{int}	internal resistance to mass transfer
R_T	total resistance to mass transfer
r	radial position within the particle
r_1	smallest solid image that can be approximated by fractal analysis
r_c	characteristic diameter of the image in fractal analysis
R	radius of spherical solid particles
t	extraction time
T	temperature
u	interstitial solvent velocity
U	superficial solvent velocity
V_c	critical volume of solute
V_T	total extraction vessel volume of a SCFE plant
Y	yield of solute
z	axial position in the bed

Greek letters

α	fraction of free solute
δ	depth of a superficial layer
ρ	density
ρ_{ap}	apparent density
ρ_s	solute density
ε	porosity of the packed bed
ε_p	porosity in a solid particle
λ	ratio of solute size to pore diameter
τ	tortuosity of the pores
μ	viscosity

Dimensionless numbers

Bi $(=k_f R/D_e K)$	Biot number for the general and LDF model
Bi $(=k_f R/d_p)$	Biot number for the DDD model
Pe $(=Ud_p/D_L)$	Peclet number
Re $(=\rho U d_p/\mu)$	Reynolds number
Sc $(=\mu/\rho/D_{12})$	Schmidt number
Sh $(=k_f d_p/D_{12})$	Sherwood number

resulted in the installation of 14 SCFE plants in China, Japan and Taiwan for a total of $\approx$35 m^3 in total extraction vessel volume in a 4-year period (1998–2001) (Fukuzato, 2003).

Effective research and development (R&D) efforts are required in Latin America to implement emerging separation technologies such as SCFE in order to take full advantage of a very abundant, diverse, and unique flora,