



Optimization of supercritical fluid extraction: Polydisperse packed beds and variable flow rates



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ABSTRACT

This theoretical study examines variable-in-time flow rates $v(t)$ and different ways of packing polydisperse ensemble of ground particles as controls in supercritical fluid extraction (SFE) to maximize the extraction yield. The so-called packing function χ is introduced to describe the local particle-size distribution in the pack along the extraction vessel. The research is based on the modified shrinking-core (SC) model for the mass transfer inside particles and assumes the pseudo-steady solvent flow in the SFE vessel. It is rigorously proven that for any variable flow rate $v(t)$ and overall particle-size distribution F , the corresponding locally-monodisperse stratified (LMS) packing χ_0 maximizes the current amount of extracted solute, while the appropriate filtration policy extends the domain of efficient particle-size distributions. Sufficient conditions that guarantee a certain extraction degree at a fixed time are deduced and formulated in terms of F -distribution. Being of obvious practical significance for finely ground substrates, optimization is shown to be rather limited for relatively big particles (commonly used in laboratory experiments), and only longer extraction times, higher oil solubility and diffusion rates allow noticeable increase in the extraction yield in this case.

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1. Introduction

Industrial technologies which use supercritical fluids become more and more popular in production of new materials, natural products, pharmaceuticals and others. One of the most attractive applications of supercritical fluids is extraction of natural products from packed beds of ground plants or other granular material – the supercritical fluid extraction (SFE) [1]. This technique gradually substitutes for traditional extraction methods all over the world, being environmentally friendly and providing selective extraction. Supercritical CO₂ is conventionally used in SFE as a solvent because it is non-toxic, non-flammable and readily available. It also has low critical temperature (~31 °C) which is crucial for extraction of thermally labile compounds (such as essential oils).

As a consequence, significant attention is drawn to understanding and optimization of the SFE process. SFE practice is usually focused on experimental tuning of extraction conditions such as solvent flow rates, temperature, and pressure in the framework of “experimental design approach” or “response surface method” to achieve the highest mass of extract at a fixed extraction time [2].

However, these research lines require a large number of experiments and do not consider more sophisticated controls which account for temporal variation of solvent flow rates and spatial inhomogeneity of particle-size distribution in a polydisperse packed bed for given thermodynamic conditions. Practical significance of such controlling factors together with other technological parameters (grinding rate and extractor dimensions) could be theoretically evaluated on the basis of an appropriate SFE model [3].

Various mass-transfer models [4–7] have been presently employed for SFE predictions and data interpretations [4–11]. Among them, the two, shrinking core (SC) [5] and broken-and-intact cell (BIC) [4], models cover a wide range of possible approaches and have shown [4,8,9,12,13] to match the overall extraction curves (OECs) with high accuracy. Based on the general description of oil diffusion in ground material, one could deduce that, in principle, the BIC model is valid in the limit of relatively small particles, when the inner part of a particle can be described in terms of spatially averaged characteristics. On the other hand, the SC model becomes more preferable in the case of relatively big particles with high oil content, when the diffusive front thickness in particles is much less than their typical size [5].

Herein we employ the modified SC modeling approach discussed [12–16] previously in detail and verified (validated) [12,13] on various available data sets.

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Nomenclature

Symbol

a	particle size (half-thickness and radius for flat and spherical particles, respectively)
A	optimal domain area
c	normalized solute concentration in the fluid phase
D	effective coefficient of oil diffusion in ground particles ($\text{m}^2 \text{s}^{-1}$)
e	bed porosity
H	vessel height (m)
f	density of overall distribution function F
F	overall particle-size distribution of original ground plant material
m_{oil}	extractable mass of the solute in the packed bed (kg)
n	particle shape factor
q	current consumption ratio
Q	total volume of solvent (m^3)
Q_{min}	minimum volume of the solvent needed to dissolve all extractable mass m_{oil} (m^3)
s	fraction of oil extracted from particles
s_1	fraction of oil extracted from particles at the end of SFE process defined by Eq. (12)
S	vessel cross-section area (m^2)
t	normalized extraction time
v	normalized variable-in-time flow rate
y	zonal oil fraction extracted from the packed bed interval $[0, z]$
Y	overall extraction curve, $Y=y(1,t)$
z	normalized spatial coordinate
UD	uniformly distributed pack
LMS	locally-monodisperse stratified pack
ILMS	inverse LMS pack

Greek symbols

α	total extraction degree, $\alpha = Y(1)$
γ	solvent consumption ratio, $\gamma = Q/Q_{\text{min}}$
δ	Dirac “delta”-function
η	solvent consumption efficiency, $\eta = \alpha\gamma^{-1}$
θ	initial density of extractable oil in ground plant material (kg m^{-3})
θ_*	equilibrium concentration of oil in the solvent (kg m^{-3})
λ	cumulative diffusion coefficient in Eq. (2)
χ	packing function, represents the local density of particle-size distribution for any given cross-section of the packed bed along the extraction vessel
ω	flow rate policy $\omega(t) = v(t)/\gamma$
ω_a	analytical approximation of optimal flow rate policy

Subscripts

0	optimal value
sc	typical scale
max	maximum value
c	the “most coarse” grinding

The paper consists of two parts. First, in Section 2, the dimensionless form of the earlier developed model is presented, and the SFE optimization problem is formulated. As mentioned above, the principal point is that original polydisperse ground plant material can be distributed along the vessel in different ways. Mathematical description of particles' packing is given in terms of the packing function χ which, by definition, represents the local density of particle-size distribution for any given cross-section of the

packed bed along the extraction vessel. Three functions: the overall particle-size distribution F of original ground plant material (or its density f), packing function χ and variable-in-time superficial velocity v (or its normalized representation – the so-called flow rate policy ω) are considered as the optimization parameters.

In Section 3, we investigate the formulated optimization problem and discuss practical importance of obtained theoretical results in case of constant flow rates as well as possible practical advantages of using variable flow rate policies.

2. SFE model and optimization problem

2.1. SFE model formulation

A generalized SFE model [12–16] based on SC approximation for the micro-scale mass-transfer phenomena inside individual particles and extended to polydisperse packed beds is employed below in our study. The convective solute-mass transport/exchange process on macro-scale level (fluid phase) is described under conventional assumption of the homogeneous flow pattern (plug flow regime [8,9]) in the bed of uniformly packed particles with constant porosity. The model has been formulated in one-dimensional presentation, in terms of the cross-sectional mean characteristics of the SFE process, in the limit of pseudo-steady (quasi-stationary) convective mass transfer [9,17] in the vessel and negligibly small axial dispersion [5,8,9,17]. Due to high diffusivity of oils in supercritical CO_2 , resistance to external oil-mass exchange between particles' surface and fluid (solvent) phase has been also neglected [16]. Model validity and practical applicability under SFE conditions typical for oilseeds (e.g. rapeseed, sunflower seed etc.) with relatively high oil content have been earlier discussed and illustrated in [12,13,15,16,18].

Aimed at mathematical investigations, we reformulate the model in dimensionless form. Hereinafter t is the normalized time, $0 \leq t \leq 1$; z – the spatial coordinate varying from 0 to 1 along the vessel, from its inlet to outlet; $v(t)$ – the dimensionless variable-in-time superficial solvent velocity and $F(a)$ – the overall particle-size distribution function (ODF) of ground plant material with the distribution density $f(a)$. By definition, $dF = f da$ is the volumetric fraction of particles with the normalized size from a to $a + da$. The particle-size distribution (functions F and f) results from grinding conditions. Let us also introduce the normalized solute concentration $c(z,t)$ in the fluid phase and the oil fraction $s(z,t,a)$ extracted from particles of size a at the moment t in the cross-section z ; $0 \leq c, s \leq 1$. It should also be noted that oil fraction s is directly linked to the shrinking core radius in spherical or flat particles [12,14,18].

The scales of the above characteristics, deduced on the basis of scale analysis and designated by subscript “sc”, are [12,18]

$$c_{\text{sc}} = \theta_*; \quad z_{\text{sc}} = H; \quad v_{\text{sc}} = \frac{Q_{\text{min}}}{St_{\text{sc}}}; \quad a_{\text{sc}}^2 = 2nDt_{\text{sc}} \frac{\theta_*}{\theta}, \quad (1)$$

where θ_* is the equilibrium concentration of oil in the solvent, θ – the initial density of extractable oil in ground plant material, H – the vessel height, S – the vessel cross-section area, $Q_{\text{min}} = m_{\text{oil}}/\theta_*$ – the minimum volume of the solvent needed to dissolve all available (extractable) mass $m_{\text{oil}} = \theta HS(1 - e)$ of the solute (oil) in the packed bed with porosity e at given temperature and pressure, D – the apparent (effective) coefficient of oil diffusion in ground particles, n – the particle shape factor ($n = 1, 3$ for flat particles of thickness $2a$ and spherical particles of radius a , respectively). The time scale t_{sc} is the duration (fixed value) of the SFE process.

Physically, the characteristic particle size a_{sc} is the boundary value which divides the ground particle ensemble in two parts of smaller ($a < 1$) and bigger ($a > 1$) particles, separating the smaller ones that can be fully extracted (at least in the limiting conditions of pure solvent, at $c \rightarrow 0$) from those that remain only partly

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