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Fractionation technologies for liquid mixtures using dense carbon dioxide.

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- Main fractionation applications include lipid mixtures, essential oils, and alcoholic beverages
- Fractionation in packed columns is the most common technology for all applications
- The manuscript characterizes packed column facilities in leading institutions
- The manuscript also discusses mixer-settlers, membrane contactors, and spray processes
- Some physicochemical properties (*e.g.*, interfacial tension) affect all technologies

## Abstract

Supercritical fluid extraction of liquid mixtures with dense CO<sub>2</sub> is an end-of-line process that is typically used to fractionate materials extracted by conventional methods. Because continuous countercurrent packed columns are the devices most commonly applied to fractionate liquid mixtures (lipid mixtures, essential oils, and alcoholic beverages), this review offers general background on the use of countercurrent Supercritical Fluid Fractionation (SFF) in packed columns. Additionally, the manuscript characterizes in detail packed column facilities in leading institutions, and describes different modes of operation of countercurrent packed columns. The manuscript also discusses less common SFF technologies for liquid mixtures such as membrane contactors, mixer-settler arrangements, and spray processes. When appropriate, the review includes extensions of these topics (*e.g.*, special uses of static mixers in SFF of liquid mixtures). In all cases, applications, future perspectives, and developments are included. Main fractionation applications include lipid mixtures, essential oils, and alcoholic beverages. Available phase equilibrium data and relevant physical properties of mixtures that are common to all technologies are also discussed. Even though a comparison between technologies is not straightforward, considerable effort has been made to identify the characteristics that make a technology more suitable for each application. In general, mixtures with low separation factors are associated with packed columns. Mixer-settler arrangements are limited to mixtures with high separation factors and when a small number of stages ( $\leq 5$ ) are required for separation. An immobile interface at the pores mouth makes aqueous systems ideal for membrane contactors, and spray processes are used when handling with high viscosity mixtures.

## Keywords

CO<sub>2</sub>, Membrane contactors, Mixer-settler, Review, Spray processes, Supercritical fractionation.

## 1. Introduction

Supercritical fluids are substances under temperature and pressure conditions above their respective critical values and where distinct liquid and gas phases do not exist. Supercritical Fluid Extraction (SFE) is a process that uses gases at high pressures as solvents to extract valuable materials. SFE has been studied to take advantage of the hybrid transport and solvent properties between gases and liquids of supercritical fluids. The most commonly used solvent in SFE is carbon dioxide (CO<sub>2</sub>), mainly because it has a near-ambient critical temperature ( $T_c$ , 31.1 °C), it is innocuous, and it is completely removable from the extract and treated substrate by simple decompression. These characteristics coupled with the selectivity of CO<sub>2</sub> towards high-value compounds in biological

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