

Membrane-based solvent extraction of aroma compounds: Choice of configurations of hollow fiber modules based on experiments and simulation

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Abstract

This work focuses on the extraction of aroma compounds by hexane in a commercial hollow fiber module. Experiments and simulations have been carried out to establish relationships between the physicochemical properties of aroma compounds and mass transport properties reached with the membrane module. The aroma compounds which have been investigated are 2-phenylethanol (PE), benzaldehyde (BA), ethyl butyrate (EB) and dimethyltrisulfide (DMTS). These four compounds have been chosen because they present a great range of partition coefficient values between water and hexane. The parameters of the process studied were the position of the feed: inside or outside the fibers and the nature of the commercial module X-30 or X-40 module by Liquicel[®]. A resistance in series model has been successfully developed to describe the mass transfer in such a system. Simulations and experiments show that the best configuration for the extraction is when the feed flows inside the shell with the X-30 module.

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1. Introduction

Numerous sectors of the food industry (fat and oil production, distilleries, seafood production, . . .) produce large amounts of effluents containing volatile molecules and aroma compounds which in general are responsible for unpleasant odors and are difficult to eliminate from effluents. Moreover, some of these odorous molecules present some interest in the food industry because they can be used as flavour compounds in flavoured food products. Indeed, food industries are greatly interested in recovering some of the aroma compounds present in their liquid effluents, because this allows fulfilling two objectives: to deodorize the effluent and to recuperate the odorous high valuable molecules in an aim to reuse them. Concerning the second aim, it is therefore necessary to implement a non-destructive extraction technique. Among extraction operations, distillation can be used

if aroma compounds are highly volatile or solvent extraction if they are hydrophobic and present a higher affinity for an organic solvent than for the aqueous effluent. Both of the traditional processes named above present some disadvantages: distillation is a quite expensive process because it is highly energy consuming and not always adapted to thermo-sensitive molecules and solvent extraction requires a high interfacial exchange area (leading often to a stable emulsion) to enhance the mass transfer. During the last 10 years, membrane technologies such as nanofiltration and reverse osmosis [1] or pervaporation [2–5] have been investigated to extract aroma compounds from aqueous solutions. Unfortunately the two first membrane processes do not offer a great selectivity and the last one is limited by low fluxes. Another alternative is to perform a membrane-based solvent extraction (MBSE), which couples the principles of solvent extraction with the high compactness and interfacial exchange area offered by hollow fiber membrane contactors (HFCs) [6]. In such a device, the membrane mainly acts as a physical barrier between the aqueous feed and the organic solvent without significant effect on selectivity. It is easy to immobilize the water–solvent inter-

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face at the pores' entrance of the membrane by applying a slight transmembrane pressure difference. The extraction is driven by a concentration gradient and transfers are not dependant on the transmembrane pressure as shown by Prasad and Sirkar [7]. HFCs offer a dispersion free contact between both phases without problems of emulsion formation, flooding or unloading [8]. This leads to a more extended choice of operating conditions when compared with classical solvent extraction because the difference between solvents density and/or similar phases velocity, which are constraints in the classical process, can be used here as operating parameters. Furthermore, both tube and shell side compartments are able to receive the feed phase. These two configurations offer different exchanging areas and different hydrodynamic conditions [9].

Of course, the use of HFCs also brings some drawbacks. The membrane induces an additional mass transfer resistance and they may be subject to shell side bypassing. Nevertheless, HFCs seem to be interesting devices to carry out aroma compound extractions as shown by different works in this area [10–12].

The aim of this paper is to find out the most appropriate module configuration to use in regards to the properties of aroma compounds. This is done by applying a double approach: coupling experiments and simulation. Experiments have been performed in a commercial HFC, on a synthetic aqueous solution containing four aroma compounds: dimethyltrisulfide (DMTS), ethyl butyrate (EB), benzaldehyde (BA) and 2-phenylethanol (PE). These compounds greatly differ in their chemical nature and therefore in their physicochemical properties such as their partition coefficient between the solvent and water. Here, we have considered hexane as a reference solvent because of its low solubility in water (9.5 mg/L at 25 °C) [13] and frequently used in the aromatic industry. The modelling of mass transfer in a commercial HFC has been developed and validated on experimental data. Simulation has been performed in order to better understand the choice of the most appropriate module configuration to use.

2. Theory

Part of this work aims to model the transfer of an organic solute from an aqueous solution towards hexane in commercial hydrophobic (polypropylene) hollow fiber membrane contactors. Two configurations have been investigated as each phase, the aqueous feed or the solvent can flow either inside the fibers or in the module's shell. In the following section, we present equations describing the configuration with the feed in the shell.

Mass transfer is described by a resistance in series model, adapted from a previous more simple model [14]. It takes in consideration four steps.

The solute:

- flows through the feed boundary layer, from the bulk phase to the interface;
- crosses the feed/solvent interface;
- diffuses in the pore filled with the solvent phase;
- flows through the solvent boundary layer.

The solute encounters three resistances to the transfer, respectively, in the feed boundary layer, in the membrane and the last in the solvent boundary layer.

The model is built with the following assumptions:

- The system works at stationary and isothermal conditions.
- Equilibrium is reached at the fluid/fluid interface.
- Each compartment (shell and fibers) is considered as ideally mixed cells in a calculus stage.
- Pore size and wetting characteristics are uniform along the whole membrane.
- The curvature of the interface does not significantly affect mass transfer, solute distribution nor interfacial area.
- Mass transfer is correctly described by the boundary layer model.
- The transport occurs only inside the membrane porosity.
- Fluids are immiscible.
- The partition coefficient of the solute is constant in the considered concentration range.
- There is only one solute to be extracted in the feed solution.

In the experimental set up, both phases are recirculated. To describe this phenomenon, we split the resolution into two successive steps. In the first step we took the assumption that the system works at stationary conditions. And then, once the overall mass transfer coefficient is determined, we take into consideration the recirculation of both phases by computing the kinetics of extraction of the aroma compounds.

The contactor is divided into elementary cells as depicted in Fig. 1. Then, it is divided into N_a stages in the axial direction, and N_r rings in the radial direction. The axial stage 1 corresponds to the entry side of the feed, the stage N_a to the inlet of the solvent. The radial stage 1 corresponds to the module's central point and the stage N_r to the periphery. As the HFC module works counter-

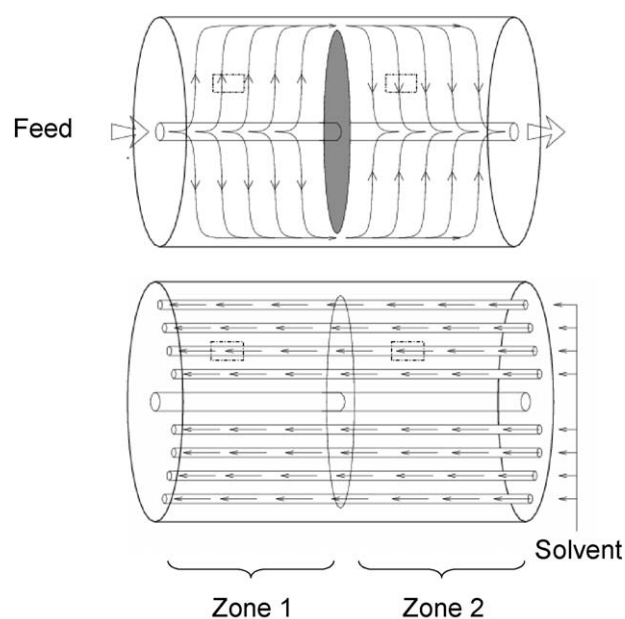


Fig. 1. Axial and radial stages considered for the modelling of the hollow fiber contactor (HFC).

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