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Liquid–liquid extraction of succinic acid using a hollow fiber membrane contactor☆

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ABSTRACT

Extraction with membrane contactors is an attractive technological alternative for organic acid separation from fermentation broths. This work aims at studying the recovery process of succinic acid, integrating both extraction and re-extraction steps. It was proposed to use a non-porous membrane, which allows more flexibility of operation without loss of efficiency. For the integrated process, a 5-fold increase in the recovery of acid from the feed stream was observed when compared to the single extraction process, leading to an overall recovery close to 50%.
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1. Introduction

The demand for organic acids produced by fermentation has grown due to their use in the manufacture of biodegradable polymers, being poly(lactic acid) the most prominent [1]. Separation is still a major cost factor: the main issue in removing organic acids from aqueous streams – as in fermentation broths – is their low concentration and the variety of substances present in the broth [2]. As an alternative to the highly expensive precipitation method currently used, several processes have been proposed, such as adsorption, ion exchange, liquid–liquid extraction and membrane separation [3–6]. Among them, membrane processes seem to be technically and economically promising. Non-dispersive solvent extraction using hollow fiber membrane contactors (also referred as membrane contactor extraction), liquid membranes and facilitated transport have been extensively studied for organic acids separation from fermentation broths [7–10].

Extraction with membrane contactors, using an organic compound as extraction agent, has many advantages over conventional liquid–liquid extraction. Membrane acts as a barrier which avoids dispersion of the liquid phases and also enhances their contact area. Moreover, the process is modular and has low energy consumption [11]. Despite all potential advantages of this process, a good extraction also relies on selecting a suitable extractant. It needs to be highly selective for the component and to have a good extraction capacity. Also, it is desirable to combine low cost with low viscosity, a moderate interfacial tension, good biological degradability and low toxicity [12]. Many works report the evaluation of hydrocarbons, alcohols, amines and phosphorous compounds as extractants for organic acids extraction. For the first two, the extraction is physical, and the distribution coefficients are small [13]. Amines form complexes with organic acids, therefore the extraction is reactive. Phosphorous compounds have been combined with amines to enhance the distribution coefficients, but high concentrations are required, increasing costs [14]. To improve the solubility of the complexes, hydrocarbons and alcohols are added as diluents to the extractant mixture. They also modify some properties of the extractants, such as viscosity [15]. Novel extractants, especially ionic liquids, have been evaluated, although only a few have presented results as good as trioctylamine (TOA), the most common extractant for organic acids extraction [16–18].

For liquid–liquid membrane extraction, porous membranes are often used, due to the lower mass transfer resistance compared to dense membranes. However, in this case, a rigorous pressure control is necessary to avoid disruption of the interface, and partially soluble extractants (as for example, primary amines) cannot be used [19]. To overcome eventual limitations of mass transfer in dense membranes, an appropriate swelling may be

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Nomenclature

$[Ext]_T$	total amine concentration in the organic phase (mol/L)
$[H_2Suc]_{aq}$	succinic acid concentration in the aqueous phase (mol/L)
$[H_2Suc]_{org}$	succinic acid concentration in the organic phase (mol/L)
K_D	distribution coefficient, defined by Eq. (1)
m_{H_2Suc}	succinic acid mass in aqueous phase (kg)
RE	re-extraction efficiency (%), defined by Eq. (2)
Recov	succinic acid recovery in membrane contactor (%), defined by Eq. (3)
Z	loading, defined by Eq. (4)

Subscripts

eq	equilibrium
ext	extraction
F	feed
o	initial
rext	re-extraction

promoted, in addition to a reduction in the membrane thickness. Lee et al. [20] already reported that extraction of solutes in liquid–liquid systems using dense membranes is feasible, but the choice of a solvent with a good interaction with the membrane material is important.

This work aims at studying the extraction of succinic acid, investigating both the extraction to the organic phase and the re-extraction into a new aqueous phase using a hollow fiber dense membrane module as a contactor. The objective is to obtain a purified acid, improving extraction efficiency. Capacity and selectivity of mixtures of 1-octanol and several amines were evaluated and the overall recovery of acid was measured for a circulating batch feed in a 2-step extraction system.

2. Experimental

To recover succinic acid (Sigma Aldrich, 99%), the extractants used were 1-octanol (Vetec), Primene JM-T® (a C16–C22 primary amine, Rohm and Haas, 80%), Primene TOA® (C8 primary amine, Rohm and Haas, 99%), *n*-Butyldiethanolamine (*n*-BDEA, Sigma Aldrich, 98.6%) and a trioctylamine (tris(2-ethylhexyl)amine, TEHA, TCI America, 93%). Because amines are viscous and corrosive, 1-octanol was used as diluent. The amine concentration in the organic phase was set in 10% w/w. All products were used as received from the manufacturers. For the membrane extraction, a cellulose diacetate membrane module (Dicea 170®, Baxter Healthcare) was selected. It was packed with dense hollow fibers, with 200 μm of inner diameter, 15 μm of wall thickness and 1.7 m^2 of contact area.

Succinic acid solutions were prepared with concentrations ranging from 0.01 to 0.5 mol/L (1–60 kg/m^3), which include the typical values found in fermentation broths [21]. Organic acid concentration in the aqueous phase was determined by HPLC (Shimadzu, SCR 102H column, H_3PO_4 as mobile phase, flow rate 0.6 mL/min, 80 °C, RI detector).

2.1. Equilibrium experiments

Equilibrium tests were performed at 25, 40 and 60 °C. Equal volumes of succinic acid aqueous solution and extractant were charged in 50 mL polypropylene vessels and stirred at 120 rpm for 1 h. The mixture was then centrifuged at 5000 × *g* for 30 min to allow complete separation of the organic and aqueous phases. Succinic acid concentration in the aqueous phase was determined by chromatography and the concentration in the organic phase was calculated from a mass balance. Distribution coefficients (K_D) were calculated, according to Eq. (1).

$$K_D = \frac{[H_2Suc]_{org,eq}}{[H_2Suc]_{aq,eq}} \quad (1)$$

where $[H_2Suc]_{org,eq}$ and $[H_2Suc]_{aq,eq}$ are, respectively, the equilibrium concentration in organic and aqueous phases.

The influence of the pH of the aqueous phase used for re-extraction on the recovery efficiency was also evaluated. Equal volumes of aqueous and organic phases (500 mL each) were charged in a vessel with controlled stirring (120 rpm) and temperature (25 °C). Succinic acid concentration in aqueous phase was 0.17 mol/L (20 kg/m^3) and the amine concentration in 1-octanol was 10%, w/w. Allowing 3 h for the equilibrium establishment, the phases were then separated by centrifugation (5000 × *g* for 30 min), and the organic phase was collected and stored. Samples were taken from the aqueous phase before and after extraction to determine the percentage of acid extracted. The pH of the re-extraction was changed by adding 0.1 mol/L hydrochloric acid or 0.1 mol/L sodium hydroxide solutions. Equal volumes of the stored organic phase and the re-extraction water were added to 50 mL polypropylene vessels. The procedure was the same as in the extraction experiments. The re-extraction efficiency (RE) was calculated as the ratio between the concentrations of acid in the aqueous phase after re-extraction and in the organic phase, as shown in Eq. (2).

$$RE\ (\%) = \frac{[H_2Suc]_{aq,eq}}{[H_2Suc]_{org,o}} \times 100 \quad (2)$$

2.2. Extraction experiments in the 2 ways membrane contactor system

For membrane contactor extraction (MCE), a system containing two coupled hollow fiber modules was set (Fig. 1), allowing simultaneously extraction of the acid from an aqueous phase (feed) to an organic phase (extractant) and re-extraction from this organic phase into a new aqueous phase (recovery), and therefore, succinic acid extraction could be achieved. This approach is expected to be very effective when the acid is extracted from a fermentation broth (a multicomponent system). In this work, tests were performed with a solution of pure succinic acid, with the purpose of evaluating the performance of the selected membrane and extractants, without interference of other solutes. The system configuration allows its operation in extraction or coupled (extraction + re-extraction) modes. Therefore, experiments were performed in both operational modes, and their results were compared.

For these tests, operation was performed with batches of succinic acid (0.17 mol/L) and extractant (10%, w/w amine in 1-octanol) solutions, and pure water for re-extraction. All solutions were allowed to recirculate. The extractant flows inside the fibers, at a flow rate of 10^{-5} m^3/s, corresponding to a Reynolds number (*Re*) of 0.4, while the aqueous phases flow externally to the fibers (succinic acid solution flows in the first module, and pure water in the second one), at a flow rate of 3×10^{-5} m^3/s (*Re* = 19). The efficiency of this process is calculated in terms of the percentage of recovery, Recov (Eq. (3)):

$$Recov\ (\%) = \frac{m_{H_2Suc,rext}}{m_{H_2Suc,F}} \times 100 \quad (3)$$

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