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Supported liquid membranes with organic dispersion for recovery of Cephalexin

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ABSTRACT

Supported liquid membranes with organic dispersion (SLM-OD) have been proposed and investigated for simultaneous removal and recovery of Cephalexin from aqueous solutions by using the carrier Aliquat 336 and a commercially available hollow-fiber module (HFM). The organic dispersion, formed by dispersing a small amount of organic membrane solution in the feed solution with a mixer, flowed through the shell side of the HFM. Investigated various parameters including organic-to-feed volume ratio, Aliquat 336 concentration, initial Cephalexin concentration, KCl concentration, shell-side organic dispersion flow rate, and lumen-side strip solution flow rate. The mass transfer mechanism of the SLM-OD technique was elucidated, and a mathematical model was developed to calculate the overall mass transfer coefficient. The results showed that the overall mass transfer coefficient increased with an increase of organic-to-feed volume ratio or increase of Aliquat 336 concentration, but reduced with the increase of initial Cephalexin concentration. Furthermore, the results indicated that the lumen and shell side flow rates had little effects on mass transfer performance and that an excess amount of KCl was necessary for the facilitated transport. The fractional mass transfer resistances of SLM-OD were calculated based on the resistance-in-series model, showing that the extraction reaction resistance was greatly reduced. In addition, the SLM-OD was shown to be superior to the supported liquid membranes with strip dispersion (SLM-SD) in terms of the improvement of mass transfer performance and the reduced volume requirement of the organic membrane solution.

1. Introduction

Cephalexin is a first-generation cephalosporin antibiotic, and it has been widely used to treat various infections due to its broad antibacterial activity [1]. The traditional production method is a 10-step chemical synthesis process which suffers from significant energy consumption, many side reactions [2] and introducing toxic solvent [3]. To overcome these shortcomings, an enzymatic synthetic method [4] has been developed and proved to be an efficient alternative for the chemical synthesis method [5]. However, the commercial application of the enzymatic method to produce Cephalexin has not been realized because of the low yield and difficulty to separate (purify) Cephalexin from product solution.

Liquid membrane (LM) is a highly integrated separation technique which combines extraction and stripping processes in one step. Since LM was firstly invented by Li in 1968 [6], various LM techniques have been proposed and extensively investigated, including bulk liquid membrane (BLM) [7], emulsion liquid membrane (ELM) [6], and supported liquid membrane (SLM) [8]. The advantages of LMs include the ease of scale-up, high selectivity, low consumption of extractant and no flooding. Therefore, LM is considered as a promising separation method for wastewater treatment, chemical and biochemical processing, and pharmaceutical and food processes [9].

LM has been proved to be an attractive technique for the separation and recovery of Cephalexin [10,11]. Sahoo et al. [10] reported the facilitated transport of Cephalexin using Aliquat 336 as the extractant in a BLM. Then, Sahoo and Dutta [11] proved that Cephalexin can be successfully separated from an aqueous solution containing 7-amino-3-desacetoxicephalosporanic acid (7-ADCA) by means of an ELM system. However, traditional LM techniques have some intrinsic shortcomings, which hinder their commercial application [12].

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In recent decades, some novel LM techniques have been proposed that offers long-term stability such as hollow fiber contained liquid membrane (HFCLM) [13], hollow fiber renewal liquid membrane (HFRLM) [14], and supported liquid membrane with strip dispersion (SLM-SD) [15]. Among them, SLM-SD has shown effective removal and recovery capabilities and has been applied in wastewater treatment [15–17] as well as separation and purification of valued organic compounds [18,19]. Vilt and Ho firstly used the SLM-SD technique to recover and concentrate Cephalexin from a dilute solution. The results showed that the recovery rate of Cephalexin was around 96–98% with the enrichment ratio ranging from 1.6 to 3.3 [18]. In subsequent work, they developed an enzymatic synthesis process coupled with the SLM-SD technique for in-situ selective removal and recovery Cephalexin using a commercially available hollow fiber module (HFM). It was found that the combination of SLM-SD and enzymatic complexation enhanced the maximum yield of Cephalexin from 32% to 42% without an enzyme deactivation problem [4].

Besides the investigation of removal efficiency and enrichment performance, the analysis of mass transfer mechanism of LM is also necessary for the future industrialization. In our previous work, the distribution of mass transfer resistances of SLM-SD for the removal and recovery of Cephalexin was estimated, and the results indicated that the extraction reaction resistance, i.e., the complexation reaction of Cephalexin and Aliquat 336, was dominant [18]. In order to decrease this resistance, a possible approach is to enlarge the mass transfer surface area between the feed and organic phases. As this mass transfer surface area in SLM-SD locates on the inner surface of hollow fibers, this would suggest that this area can be augmented by increasing the number of hollow fibers. But this would enlarge the size or number of hollow-fiber membrane modules and the amount of organic phase, resulting in an increase of the cost. In addition, from the environmental and economic perspectives, the amount of organic phase should be limited. Actually, in the solvent extraction and HFRLM techniques, the mass transfer area can be greatly increased by dispersing the organic solution in the aqueous solution. In a HFRLM technique, the dispersion flowed through the lumen side of HFM, and the organic-to-aqueous volume ratio was normally less than 1:10, which enhanced the mass transfer performance [14].

In this work, the SLM with organic dispersion (SLM-OD) technique was proposed and demonstrated for the removal and recovery of Cephalexin from its dilute aqueous solutions. Organic dispersion, formed by dispersing the organic membrane solution in the feed solution with a mixer, flowed through the shell side of a HFM. A series of important experimental parameters were investigated, including the organic-to-feed volume ratio, Aliquat 336 concentration, initial Cephalexin concentration, KCl concentration, and lumen- and shell-side flow rates. This allowed for identifying the critical parameters. A mathematical model was developed to determine the overall mass transfer coefficient of SLM-OD. Furthermore, according to the resistance-in-series model, the distribution of mass transfer resistances of the SLM-OD system was calculated. This showed a significant reduction of the mass transfer resistance due to the extraction reaction for the SLM-OD vs. the supported liquid membranes with strip dispersion (SLM-SD). In addition, the SLM-OD was shown to be superior to the SLM-SD in terms of the reduced volume requirement of the organic membrane solution.

2. The SLM-OD technique

The SLM-OD technique is illustrated schematically in Fig. 1, in which a small amount of organic membrane solution is dispersed as droplets in the continuous aqueous feed solution by a mixer, while in the SLM-SD technique, the dispersed phase is the aqueous strip solution. Typically, a hollow-fiber module (HFM) containing

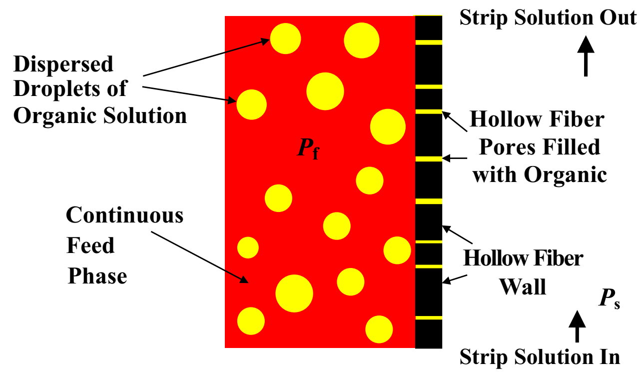

Fig. 1. A schematic diagram of an enlarged view of the SLM-organic dispersion (SLM-OD) process.

microporous hydrophobic polymer hollow-fibers should be used to obtain a large mass transfer area. In a HFM, the dispersion phase can flow on the shell side while the aqueous strip phase can be on the lumen-side. In order to prevent any sipping of the organic membrane solution, the pressure of the feed phase (P_f) containing the organic dispersion and the aqueous strip phase pressure (P_s) should be approximately the same (about 34.5 kPa, 5 psi). In this work, the organic dispersion was pumped through the shell side of the HFM.

3. Theory

3.1. General transport mechanism

In SLM-OD, the organic membrane solution is dispersed in the continuous aqueous feed solution. The multiphase flow behavior is different from that of SLM-SD, suggesting that SLM-OD may have a different transport mechanism, which is described as follows:

- Solvent extraction phenomenon in the organic dispersion vessel
  Due to the direct mixing of the feed and organic phases, a small amount of target species in the feed phase can be quickly extracted into the organic droplets, which are a small fraction in the dispersion.
- Mass transfer on the shell side of the module
  On the shell side of HFM, the dispersed organic droplets and the outer surface of the hollow fibers provide a large mass transfer area at the feed/organic interface. There are three types of transport processes.
  a. At the feed/organic interface located on the outside diameter of each hollow fiber, the target species reacts with the carrier to form the solute–carrier complex and is partitioned into the organic phase, i.e., the supported liquid membrane phase.
  b. At the feed/organic interface located on the surface of each organic droplet, the target species reacts with the carrier to form the solute–carrier complex in the organic droplet. Due to the small size of each organic droplet in the dispersion, the concentration profile of the solute–carrier complex in the organic droplets is assumed to be uniform. The pumping flow action enhances the fulfillment of this assumption.
  c. The solute-loaded droplets may collide with the hollow fibers due to the effect of the flow field. In this process, the complex in the droplets may be directly transported to the feed/organic interface located on the outside diameter of hollow fibers. This process may bring about the mass transfer intensification.
- Mass transfer in the membrane supported in pores
  Because the microporous hydrophobic polymer hollow-fibers have been wetted by the organic membrane phase, the complex can then diffuse across the membrane supported in the porous wall of each hollow fiber.