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Modeling study on the mass transfer of hollow fiber renewal liquid membrane: Effect of the hollow fiber module scale

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ABSTRACT

The effect of hollow fiber module scale on the mass transfer performance of hollow fiber renewal liquid membrane technique is studied experimentally and theoretically. Five scales of polypropylene hollow fiber membrane modules with same packing density and effective length are used for experiments. $CuSO_4$ aqueous solution is used as feed phase, the organic solution of LIX984N in kerosene is used as liquid membrane phase, and H_2SO_4 is used as the stripping phase. The non-ideal flow status on the shell-side of the hollow fiber module is described by residence time distribution curves and quantitatively characterized by the Peclet number. Peclet number increases with the increase of L/d in hollow fiber module, and reaches its maximum value when L/d is 20, then decreases. The overall mass transfer coefficient is directly proportional to the Peclet number on the shell side in single-pass, recycling and cascade operation modes in the hollow fiber renewal liquid membrane process. A new mass transfer correlation is proposed to quantitatively describe the non-ideal flow on the shell-side of the hollow fiber module with Peclet number. Then, the corresponding mathematical model for hollow fiber renewal liquid membrane process is developed and the calculated results show good agreement with the experimental data.

1. Introduction

Hollow fiber membrane modules have a very broad range of applications in mass transfer and separation processes [1], such as filtration, gas–liquid membrane absorption, pervaporation, membrane distillation, membrane extraction, etc. which are used as separators, contactors and reactors [2].

Hollow fiber membrane modules are generally designed as a contactor with shell-and-lumen structure, similar to a lumen and shell heat exchanger. A bundle of porous hollow fibers are packed randomly in the cross-section of the hollow fiber membrane module. In such modules, a surprisingly high interfacial area per volume can be achieved over conventional equipment; no loading and flooding happens as in the conventional column contactors; entrainment is eliminated because of no direct phase-to-phase contact; operation is easy to handle within a wide range operation region; and some other advantages are reported in literatures [1,3,4]. It is worth taking into account the liquid membrane process in hollow fiber module. As a simultaneous extraction and stripping process, it has attracted more attention recently due to its potential advantages [5].

However, the liquid membrane process in hollow fiber module encounters many problems, especially the uncertainty of process design and scale-up caused by non-ideal flowing on the shell side, which obstructs the industrial application of liquid membrane technique in hollow fiber module. The existing mathematical models for mass transfer have serious limitation to get valid results for liquid membrane process in hollow fiber module. The structure and scale of the hollow fiber module have significant influences on the performance of flowing status and mass transfer efficiency. The random packing of fibers on the shell side makes the fiber distribution non-uniform, which will lead to severe fluid channeling and bypassing on the shell side of the module. Therefore, the mass or heat transfer process is compromised [6,7].

In a design process, the detailed mass transfer parameters in hollow fiber module are necessary for a target process. The mass transfer performance in hollow fiber module, such as gas-liquid absorption, liquid–liquid extraction, is often described by *resistance-in-series* model, which includes resistances of diffusion across membrane pores and the boundary layers of the flowing fluids on both the lumen side and shell side [1,8]. The estimation of the individual mass transfer coefficient on the lumen side and the membrane phase has been developed extensively with agreements to experimental results [9,10]. Most mass transfer correlations on

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Table 1
Mass transfer empirical correlations for mass transfer on the shell side.

References	Correlation	Packing density
Yang and Cussler [3]	$Sh = 1.25\frac{(1-\phi)^{1.79}}{\phi^{0.86}}\left(\frac{d_{out}}{L}\right)^{0.93} Re_w^{0.93} Sc_w^{0.33}$	0.03, 0.26
Prasad and Sirkar [11]	$Sh = 5.8\frac{(1-\phi)^{2.2}}{\phi^{0.6}}\left(\frac{d_{out}}{L}\right) Re_w^{0.6} Sc_w^{0.33}$	0.04, 0.197, 0.40
Costello et al. [12]	$Sh = (0.53-0.58\phi)\left(\frac{(1-\phi)}{\phi}\right)^{-0.47} Re_w^{0.53} Sc_w^{0.33}$	0.319 to 0.758
Bao et al. [13]	$Sh = 1.38(2.35\phi-0.07)\left(\frac{1-\phi}{\phi}\right)^{1/3}\left(\frac{d_{out}}{L}\right)^{1/3} Re_w^{1/3} Sc_w^{1/3}$	0.06 to 0.75
Asimakopoulou and Karabelas [14]	$Sh = 1.45\left(\frac{d_{out}}{L}\right)^{1/3} Re_w^{1/3} Sc_w^{1/3}$	0.05 to 0.45

the lumen side are based on Graetz–Leveque correlation, especially for the laminar flow. Unfortunately, the mass transfer empirical correlations on the shell side proposed by researchers have larger differences as listed in Table 1 [3,11–14], mainly because of uncertainties of flowing status caused by the configuration of hollow fiber membrane module, such as scale and structure of module, fiber packing, bundle geometry, as reviewed by Gabelman and Hwang [1]. Then, there is an urgent need to find a general quantized parameter to describe these effects on the process, therefore, to improve its mass transfer performance.

Hollow fiber renewal liquid membrane (HFRLM) is a new simultaneous extraction and stripping technique with good stability and high mass transfer rate proposed by Zhang et al. [15,16]. The aim of this work is to study the effect of hollow fiber membrane module scale on the mass transfer performance of HFRLM process. In experiments, $CuSO_4$ aqueous solution is used as feed phase, the organic solution of LIX984N in kerosene is used as liquid membrane phase, and H_2SO_4 is used as the stripping phase. Five scales of the hollow fiber membrane modules with identical fiber packing density and various shell diameters are tested. The residence time distribution (RTD) curves are measured to explore the shell-side flow status. The mass transfer experiments are conducted with single-pass, recycling and cascade modes to indicate the effects of non-ideal flow on the mass transfer performance. The mathematical model of mass transfer for HFRLMs process based on the resistance-in-series model and mass balance law is proposed by using Pelect number to describe the non-ideal flow in the shell side.

2. Experimental

2.1. Reagents

LIX984N, from Cognis Ltd. Co., is a mixture of Lix860N and Lix84I with 1:1 (v/v). Copper sulfate anhydrous, from Shanghai Tingxin chemical reagent plant, is an analytical grade reagent with a purity $>99.0\%$. Commercial kerosene, from Tianjin Damao chemical reagent plant, is washed twice with 20% (vol.) H_2SO_4 to remove aromatics and then three times with deionized water. All other chemicals are of analytical grade and without further purification except for kerosene. The feed phase is prepared by dissolving a weighed amount of $CuSO_4$ in deionized water and the pH is adjusted with H_2SO_4. The organic phase (i.e. liquid membrane phase) is prepared by dissolving of 10% LIX984N in kerosene.

2.2. Apparatus

Five hollow fiber membrane modules are used in this work with identical fiber packing density (0.3) and effective length (30 cm), which are self-assembled with different shell diameter specifically designed for this experiment. The used polypropylene (PP) hollow fibers are from Qiushi Ltd. Co (Hangzhou, China). Additional information about these modules is provided in Table 2.

Table 2
Characteristics of the hollow fiber modules.

NO.	Effective length of module, L (m)	Inner diameter of module, D_i (m)	Number of fiber, n	Outer diameter of fiber, $d_o \times 10^4$ m	Thickness of fiber, $\delta \times 10^4$ m
1#	0.3	0.0080	95	4.5	0.5
2#	0.3	0.0120	213	4.5	0.5
3#	0.3	0.0150	333	4.5	0.5
4#	0.3	0.0205	592	4.5	0.5
5#	0.3	0.0250	1000	4.5	0.5

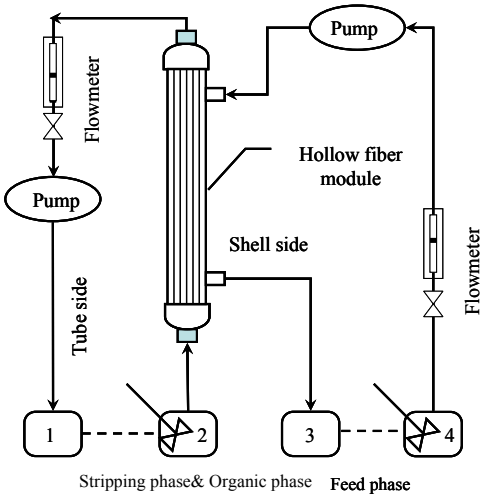

Fig. 1. Experimental set-up of the HFRLM process.

2.3. Experimental procedures

In order to investigate the flow status on the shell side of the hollow fiber membrane module, RTD curves are detected by the pulse tracer input technique using saturated KCl solution. The deionized water is pumped through the shell side of the module. The quantitative saturated KCl aqueous is injected in the inlet fluid through the shell side. The lumen side and the pores of the fibers are both filled with kerosene to prevent the leakage of KCl aqueous into the lumen side. The concentration of KCl in outlet fluid on the shell side is monitored online by electric conductivity meter.

The experimental set-up of HFRLM process is shown in Fig. 1. Prior to the experiments, the hydrophobic fibers are pre-wetted with organic phase more than 0.5 h in order to make the pores of fibers fully filled with organic phase. The stirred mixture of the stripping phase (2.0 mol $L^{-1}H_2SO_4$ aqueous solution) and the organic phase at a volume ratio of 20:1 is pumped through the