Separation and Purification Technology 148 (2015) 25–31

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Diffusion dialysis of sulfuric acid in spiral wound membrane modules: Effect of module number and connection mode

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ARTICLE INFO

Article history:
Received 15 February 2015
Received in revised form 22 April 2015
Accepted 23 April 2015
Available online 9 May 2015

Keywords:
Sulfuric acid
Diffusion dialysis
Spiral wound module
Acid recovery
Integrating system

ABSTRACT

The main goal of this work is to explore the transfer of sulfuric acid in spiral wound membrane modules. The effect of various parameters such as connection modes, number of membrane modules, flow rate and initial feed concentration are fully investigated. The results show that the acid recovery ratio increases with the number of membrane modules and decreases with the flow rate but does not apparently changes with the initial feed concentration. Meanwhile, both the recovered acid concentration and the dialysate acid concentration are directly proportional to the feed concentration for different number of SWDD modules. For double SWDD membrane module system, the mode D-d (series connection in both acid side and water sides) seems to be the best one from the integrated viewpoints of acid recovery ratio and recovered acid concentration. For multi-modules system, the treating capacity is much higher than the sum contributed from each single module.

1. Introduction

Sulfuric acid is known to be one of the key inorganic acid and has been widely used in chemical reaction and metal industries for surface treatments such as electrolysis, electroplating and acid pickling because of its relative inexpensiveness in comparison with other inorganic acids [1,2]. During these processes, large amounts of waste sulfuric acid solution are generally generated. Direct disposal of the waste solution will not only squander lots of recovery resource but also result in environmental contamination [3]. It is thus highly recommended to develop an alternative suitable method to dispose the waste solution and recover sulfuric acid therein. At present, several methods are available to treat the acidic waste solutions including neutralization with alkalis, concentration with evaporation and separation with membrane-related technologies [1,4–9]. Among all these available methods, diffusion dialysis (DD) provides an attractive acid recovery method from the view point of energy saving, low-cost, and environment-friendly nature [10,11].

DD is a spontaneous diffusion process which not only relies on the concentration difference but also needs no external driving forces and additives. Plate-and-frame is known to be a common-used membrane module in diffusion dialysis process. However, the low mass treating capacity and mass transfer efficiency limits its large scale applications. Compared with plate-and-frame membrane module, spiral wound diffusion dialysis (SWDD) membrane module has gained much attention and possesses many advantages like compact equipment, small volume, large handling capacity, convenient assembling and transportation, sufficient mass transfer [10], etc. Recovering hydrochloric acid from wastewaters containing aluminum ions by SWDD membrane module has been reported in recent papers [10,12]. The work committed itself to the investigation of single SWDD membrane module and monadic acid. Till date, the mass transfer of dibasic acid in SWDD membrane module is not highly explored and is an important issue to extend the application of SWDD in various industrial fields. In addition, module connection modes and number has never been investigated in SWDD processes.

Therefore, the main goal of this paper is to study the mass transfer of sulfuric acid in SWDD membrane modules. The effect of operating variables such as flow rate, membrane module connection modes and number, initial feed concentration on diffusion performance was fully investigated.

2. Experimental

2.1. Materials

The feed used for the diffusion dialysis experiment was prepared from the analytical pure sulfuric acid. The anion exchange

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http://dx.doi.org/10.1016/j.seppur.2015.04.033
1383-5866/

Nomenclature

S	single SWDD membrane module
D	double SWDD membrane module
T	triple SWDD membrane module
C_d	concentration of H^+ in dialysate
C_r	concentration of H^+ in recovered acid
C_f	concentration of H^+ in feed
$C_{d,\ D\text{-}a}$	dialysate concentration of H^+ in connection mode D-a
$C_{d,\ D\text{-}b}$	dialysate concentration of H^+ in connection mode D-b
$C_{d,\ D\text{-}c}$	dialysate concentration of H^+ in connection mode D-c
$C_{d,\ D\text{-}d}$	dialysate concentration of H^+ in connection mode D-d
$C_{r,\ D\text{-}a}$	recovered acid concentration of H^+ in connection mode D-a
$C_{r,\ D\text{-}b}$	recovered acid concentration of H^+ in connection mode D-b
$C_{r,\ D\text{-}c}$	recovered acid concentration of H^+ in connection mode D-c
$C_{r,\ D\text{-}d}$	recovered acid concentration of H^+ in connection mode D-d
$C_{r,\ S}$	recovered acid concentration of H^+ in connection mode S
$C_{r,\ T}$	recovered acid concentration of H^+ in connection mode T
Q_d	flow rate of dialysate
Q_r	flow rate of recovered acid
Q_f	flow rate of feed
Q_w	flow rate of water
Q	flow rate
R	total acid recovery ratio
R_T	acid recovery ratio of connection mode T
$R_{D\text{-}d}$	acid recovery ratio of connection mode D-d
R_S	acid recovery ratio of connection mode S
$J_{H_2SO_4}$	molar flux of sulfuric acid through the membrane
A	total membrane area
w	width of interval channel
l	length of the membrane
K	overall mass transfer coefficient
n_r	molar of sulfuric acid through the membrane
V_r	volume of recovered acid
t	time
ΔC	concentration difference across the membrane
k_d	mass-transfer coefficients in feed compartment
k_r	mass-transfer coefficients in recovery acid compartment
k_m	mass-transfer coefficients within the membrane
R_h	mean hydraulic radius
S	cross-sectional area of the interval channel circularity
Z	wetted perimeter
d_1	external diameter of the interval channel circularity
d_2	internal diameter of the interval channel circularity
Re	Reynolds number
h	height of interval channel
ρ	density of solution
μ	viscosity of solution
u	average linear velocity in the interval channel

membrane (DF120) used in the SWDD membrane module was provided by Shandong Tianwei Membrane Technology Co., Ltd., China, and its main characteristics were: ion exchange capacity (*IEC*) was 1.82 mol/kg, water content (W_R) was 42.34%, membrane area resistance (*MER*) was 2.7 Ω cm^2 (25 °C). Distilled water was used all throughout the experiment.

2.2. Experimental apparatus

The experimental set-up used in this study is shown in Fig. 1a–f. The SWDD membrane module was made by our own laboratory. The detailed description of its structure was described thoroughly in our previous patent and papers [10,11,13]. In brief, the membrane area for one module is 1.2 m^2 with an effective membrane area around 1.0 m^2. The module length is 470 mm and its section radius is about 50 mm. Scheme of SWDD membrane module is shown in Fig. 1g. Herein feed solution and water flow at the two different sides of membrane respectively and the two different solutions were kept on the condition of spirally radial counter current. As an example shown in Fig. 1e, feed enters into the SWDD membrane module from feed tank to the dialysate tank while water enters the module from water tank to the recovered tank. Before the peristaltic pumps were turned on, two interval channels of SWDD membrane module were rinsed by 1.5 L of water and 1.5 L of feed in order to eliminate the gas bubbles inside the membrane module. The flow rate ratio of feed to water was 1.0 and the flow rate ranges from 0.36 to 3.6 L/(m^2 h). The initial feed concentration was 0.2–4.0 mol/L of sulfuric acid. The peristaltic pumps were used to regulate the feed and water flow rate were provided by Baoding Longer Precision Pump Co., Ltd., China. All the experiments were conducted at ambient temperature around 20 °C.

2.3. Analyses and data calculations

The representative samples in the dialysate and recovered acid solution were taken to analyze with pre-determined time interval respectively. The concentration of H^+ was determined by titration with a calibrated Na_2CO_3 solution with methyl orange as an indicator. All the experimental data were collected through three independent measurements.

The mass transfer performance of acid for SWDD membrane module was characterized by the total acid recovery ratio *R*, which is calculated from the following formula [10,14]:

$$R = \frac{C_r Q_r}{C_f Q_f} \times 100\% = \frac{C_r Q_r}{C_r Q_r + C_d Q_d} \times 100\% \tag{1}$$

where C_d, C_r, and C_f are the concentration of H^+ in the dialysate, recovered acid and feed, respectively. Q_d, Q_r, and Q_f are the flow rate of the dialysate, recovered acid and feed respectively. The values of C_d and C_r for calculating *R* were determined at the time when the diffusion processes were stable.

3. Results and discussion

3.1. Determination for flow type in SWDD

As described in the literature [15], the diffusion mass transfer performance was mainly influenced by the fluid flowing condition in the interval channel. Thus the Reynolds number (*Re*) was taken into account. It was calculated on the basis of noncircular tubes by employing a mean hydraulic radius (R_h) as shown in Eqs. (2)–(5) [16,17].

$$R_h = \frac{S}{Z} \tag{2}$$

where R_h is the mean hydraulic radius, S the cross-sectional area of the interval channel circularity, and *Z* the wetted perimeter. They are defined as:

$$S = \pi\left(\frac{d_1}{2}\right)^2 - \pi\left(\frac{d_2}{2}\right)^2 \tag{3}$$