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High-efficiency HFSLM for silver-ion pertraction from pharmaceutical wastewater and mass-transport models

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ABSTRACT

Hollow fiber supported liquid membrane (HFSLM) is a favorable technique for the pertraction of metal ions, especially at very low metal concentration. In this work, the pertraction of silver ions from acidic pharmaceutical wastewater via HFSLM was investigated. Pharmaceutical wastewater containing 30 mg/dm^3 of silver ions and 120 mg/dm^3 of ferric ions was subjected to HFSLM as a feed solution. LIX 84-I dissolved in organic solvent together with $Na_2S_2O_3 \cdot 5H_2O$ solution was selected for use as a liquid membrane and a receiving solution, respectively. The influence of ferric ions on the pertraction of silver ions was studied firstly using wastewater with normal ferric ion concentration and secondly using wastewater with ferric ion precipitation by phosphoric acid solution. The highest pertraction of silver ions was achieved by using 0.1 M of LIX 84-I and 0.5 M of $Na_2S_2O_3 \cdot 5H_2O$ solution at pH of feed and receiving solutions of 3.5 and 2. The flow rates of feed and receiving solutions were 0.2 dm^3/min. 0.6 mg/dm^3 of silver ions that remained in the wastewater was below the mandatory discharge limit. No effect of normal ferric ion concentration in the wastewater on silver ion pertraction was observed. The crucial parameters were defined to confirm the efficiency and reliability of the system. Finally, the controlling transport regime of silver ion pertraction across HFSLM was determined by the diffusion flux and reaction flux models.

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Keywords: HFSLM; Silver ions; Pertraction; Pharmaceutical wastewater; Mass transport model; Separation

1. Introduction

The contamination of silver in the environment is normally found in wastewater from the medical and photographic-imaging industry, electronics industry and the manufacturing of silverware and jewelry. Silver compounds and free ions are toxic (Altin et al., 2010; Chaudry et al., 2008). The concentration of silver ions of only 1–5 mg/L can kill aqueous organisms, insects, trout and flounder. The accumulation of silver in plants and animals leads to adverse effects on their growth but in humans it leads to convulsion and an argyria disease. This disease is an unusual illness marked by discoloration of skin, nails, mucous tissues and organs (Wijnhoven et al., 2009; Sue et al., 2001). Due to its toxicity, the removal of silver from wastewater is of paramount importance.

In compliance with the mandatory discharge limit (1.0 mg/L) (Announcement of the Industrial Estate Authority of Thailand, 1998), common techniques have been used to remove silver from wastewater such as electrolysis, chemical precipitation, ion exchange and adsorption. Nevertheless, these methods are not sufficiently effective for the removal of silver. Electrolysis requires high current density, a long operating time and pH adjustment approaching 90% of removal. In addition, its removal efficiency is lower when it operates

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Nomenclature

A	effective surface area of the hollow fiber (dm^2)
A_c	cross-sectional area of the hollow fiber (dm^2)
b	parameter in Eq. (25)
d_i	diameter of the hollow fiber
E	removal efficiency
F	parameter in Eq. (3)
Gz	Graetz number
i, j	integer numbers
J	flux of silver ions (mg/dm^2 min)
K_{ex}	equilibrium constant (dm^3/mg)
k_f	mass transfer coefficient of silver ions in the feed phase (dm/min)
k_m	mass transfer coefficient of complex species across liquid membrane phase (dm/min)
k_r	reaction rate constant (min^{-1})
L	length of a hollow fiber (dm)
M	memory effect
M_w	molecular weight of water (mg/mol)
m	parameter in Eq. (25)
n	reaction order of removal reaction
n_f	silver ion amount in the feed phase (mg/dm^3)
n_r	silver ion amount in the receiving phase (mg/dm^3)
n_w	silver ion amount the wastewater after contact with the liquid membrane phase (mg/dm^3)
P	pertraction efficiency
P	number of repeated analysis
Q	volumetric flow rate of feed solution (dm^3/min)
R	mass transfer resistance (min/dm)
RSD	relative standard deviation
r_{-Ag^+}	reaction rate of silver ion removal (mg/dm^3 min)
r_i	inside radius of the hollow fiber (dm)
r_o	outside radius of the hollow fiber (dm)
SD	standard deviation
Sh	Sherwood number
T	temperature of the HFSLM system (K)
t	removal time (min)
V	volume (dm^3)
υ	linear velocity of the feed solution (dm/min)
x	direction of the hollow fiber axis (dm)

Greek letters

α	a constant in Eq. (34)
β	a constant in Eq. (36)
λ	parameters for β in Eq. (36)
ε	porosity of the hollow fibers (%)
τ	tortuosity of the micro pores of hollow fibers
τ_0	a constant in Eqs. (34)–(36)
η_w	viscosity of water (cP)
σ	variance within the same sample

Symbols

[]	concentration
⟨⟩	average value
—	species in liquid membrane phase or difference of outlet concentrations of silver ions as considering at time t and initial time in Eqs. (34)–(36)
ϕ	association factor of water
D	diffusion coefficient of silver ions (dm^2/min)
υ_{Ag^+}	molar volume of silver ions at normal boiling point

Subscripts

Ag^+	silver ions
Expt.	experimental results
f	feed phase
f, in	in inlet concentration of silver ions in the feed phase
f, out	out outlet concentration of silver ions in the feed phase
fi	interface between feed and liquid membrane phases
m	liquid membrane phase
ri	interface between liquid membrane and receiving phases
Theo.	modeled results
w	water
0	initial concentration

over extending time due to the precipitation of sulfide on the cathode. Chemical precipitation cannot achieve the mandatory discharge limit because of the difficult precipitation of silver compounds in the wastewater (Othman et al., 2006). Ion exchange needs to regenerate the ion-exchange resin and consumes extra chemicals (Vernekar et al., 2012). Besides, nonselective adsorption using common adsorbents is a main drawback and results in difficult and expensive for reuse (Lam et al., 2008).

To over come of these problems, liquid pertraction technologies have been accomplished to remove and recover metal ions from such diluted solutions. One promising technique of liquid pertraction technologies is the use of supported liquid membrane (SLM). This technique has a lot of advantages over the common techniques, such as low capital and operating costs, low energy consumption and easy operation (Lv et al., 2007; Yang et al., 2007). The SLM technique is a three-phase system in which the liquid membrane phase separates between the feed and receiving phases. Hollow fiber supported liquid membrane (HFSLM) is designed based on the SLM technique. It is found to be highly proficient in low-level metal removals that can be shown by some examples of previous works in Table 1. Other outstanding advantages of HFSLM are high transport flux, no entrainment and flooding, very large interfacial area, simultaneous removal and recovery steps in one single stage etc. (Markoš, 2011; Seidl et al., 2007; Rathore et al., 2001; Valenzuela et al., 1999).

In general, the pertraction of metal ions by HFSLM depends on the type and concentration of a single carrier or types and concentration ratio of the synergistic carriers; type of organic solvent to dissolve the carrier; type and concentration of the receiving solution; concentration of metal ions in the feed solution; pH, flow rates and flow patterns of feed and receiving solutions. As a matter of fact, the pertraction of metal ions across HFSLM involves the diffusions – metal ions across the feed phase, complex species from the chemical reaction

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