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Separation of amoxicillin using trioctylmethylammonium chloride via a hollow fiber supported liquid membrane: Modeling and experimental investigation

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ABSTRACT

The separation experiments of amoxicillin via hollow fiber supported liquid membrane (HFSLM) were performed under various operating conditions to find the optimal parameters i.e. pH, feed concentration, carrier concentration, and flow rates of feed and stripping solution. Percentages of extraction and recovery of amoxicillin from the feed phase reached 85.21% and 80.34%, respectively. The aqueous-phase mass transfer coefficient (k_f) and organic-phase mass transfer coefficient (k_m) were reported to be 3.57×10^{-2} and 0.70×10^{-2} cm/s, respectively. Furthermore, a mathematical model was developed to predict the concentration of amoxicillin at different times. The results showed promising agreement with experimental data.

Introduction

Amoxicillin, 6-(*R*-hydroxy-a-amino phenyl acetamido) penicillanic acid, is the only phenolic penicillin which is used as an antibacterial drug [1]. Amoxicillin is a β-lactam antibiotic that has a broad in vitro spectrum against gram negative and gram positive bacteria, as well as good absorption and penetration into tissues. It is a kind of frequently used antibiotic to treat many kinds of infections [2]. The molecular weight of amoxicillin is 365 and it is soluble in water. The molecular structure of amoxicillin is shown in Fig. 1(a). The presence of amoxicillin and other kinds of antibiotics in the environment is of concern due to their potential to promote bacterial resistance [3] as well as trigger long term adverse human health effects. Chemical disinfection which is one of the essential water treatment processes may aid in their removal from industrial wastewater but may also form by-products that can remain biologically active. Amoxicillin wastes can cause unpleasant odor, skin disorder, and may cause microbial resistance among pathogen organisms or the death of microorganisms which are effective in wastewater treatment [4]. The resistant bacteria may cause disease that cannot be treated by conventional antibiotics. For these reasons, amoxicillin waste needs to be treated before being disposed to the environment.

There are many methods for the removal of amoxicillin from wastewater, such as sand filtration [5], chemical coagulation or flocculation [6], chlorination [7], ultraviolet (UV) radiation [8], ozonation and advanced oxidation processes (AOP), adsorption and membrane process [9]. Techniques that have been gaining attention in the past few years are membrane process and liquid membrane process. Hollow fiber supported liquid membrane (HFSLM) is a system based on the liquid membrane process and membrane process. HFSLM technique has specific characteristics of simultaneous extraction and stripping processes of low concentrations of target species in one single stage [10]. Some other advantages of the hollow fiber contactor over traditional

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Nomenclature

C	concentration (mmol/L)
k_f	aqueous feed mass transfer coefficient (cm/s)
k_m	organic mass transfer coefficient (cm/s)
K_{ex}	extraction equilibrium
K_{st}	stripping equilibrium
D	distribution ratio
P	permeability coefficient (cm/s)
P_m	membrane permeability (cm/s)
A	effective area of hollow fiber (cm^2)
V_f	volume of feed phase (cm^3)
r_m	the log-mean radius of the hollow fiber (cm)
r_i	internal radius of the hollow fiber (cm)
r_o	external radius of the hollow fiber (cm)
Q	volume metric flow rate (cm^3/s)
L	length of the hollow fiber (cm)
N	number of hollow fiber in module
R_m	organic membrane mass transfer resistance (s/cm)
R_i	aqueous mass transfer resistance (s/cm)
t	time (min)
J	flux (mol/cm^3/min)
n	order of extraction reaction

Greek letters

τ	tortuosity of membrane
ε	porosity of membrane

Subscripts

f	feed phase
s	stripping phase
m	membrane phase
i	inter phase
0	initial concentration

separation techniques include lower capital and operating costs [11], lower energy consumption [12], less solvent used and high selectivity [13]. These advantages render the HFSLM system more suitable for the treatment or separation of amoxicillin.

The present work studies the effects of parameters which influence the effective separation of amoxicillin by using HFSLM technology based on trioctylmethylammonium chloride (Fig. 1(b)) as an extractant. In addition, the experimental data of amoxicillin concentrations in the outlet feed solution were compared with the results from the mathematical model based on HFSLM batch separation system.

Theoretical background

Transport mechanism and separation of amoxicillin

In a hollow fiber supported liquid membrane (HFSLM) system, the supported liquid membrane is embedded in an organic extractant which separates the feed from the stripping phases. At the feed–membrane interface, the amoxicillin reacts with the extractant to form complex species. Subsequently, the complex species diffuse across the liquid membrane to react with the stripping solution at the membrane–stripping interface. Then, they are stripped into the stripping phase. The separation of amoxicillin was studied using the extractant trioctylmethylammonium chloride (aliquat 336 Cl) and involves their facilitated coupled counter-transport across HFSLM that is illustrated in Fig. 2.

The mechanism using the extractant is described in the following steps:

Step 1 Amoxicillin (Amox) in the feed phase is transported to the interface between the feed and liquid membrane phases. Then, it reacts with the extractant ($\overline{QCl}$) to form the complex species ($\overline{QAmox}$) and releases chloride ions into the feed phase.

Step 2 The complex species diffuses to the interface between the liquid membrane and receiving phases by its concentration gradient.

Step 3 The complex species reacts with the receiving solution to recover amoxicillin into the receiving phase while the extractant transports back to the opposite side and reacts again with the amoxicillin in the feed phase. Thus, both amoxicillin and chloride ions are counter-transported across HFSLM.

In short, when the diffusion steps in the facilitated transport are slower than the chemical reaction steps, the transport is described as the diffusion transport regime [14,15].

In developing the reliable mathematical model in order to describe the transport mechanism of the target species across liquid membrane, general models based on diffusion or chemical reaction were taken into consideration [16,17]. The diffusional process through a liquid membrane is affected by the porosity and tortuosity of the polymeric support [18]. The basic parameter of the transport regime to determination of transport parameters was the diffusion coefficient [18–20]. The type of movements associated with the migration of the complexes through the organic phases of the liquid membrane was indicated by the diffusion coefficient [19–21].

1. The diffusion coefficient in the range of 10^{-6}–10^{-5} cm^2 s^{-1} indicated movement of pure diffusion for a very stable complex in an organic phase [21].

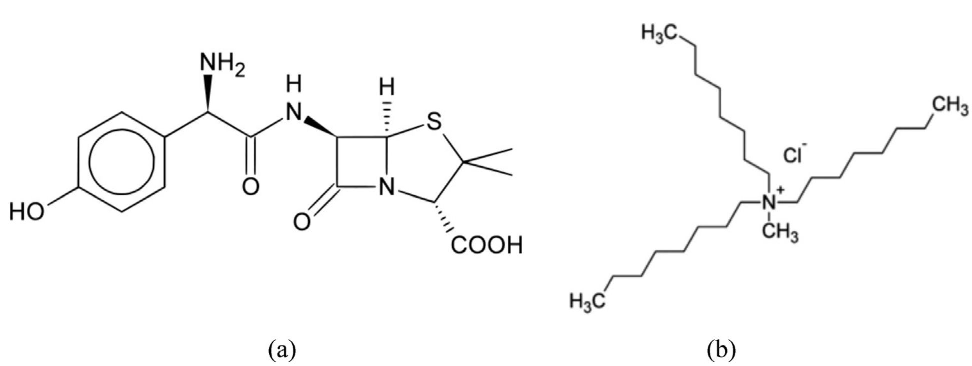

Fig. 1. The structures of (a) amoxicillin and (b) trioctylmethylammonium chloride (aliquat 336).