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The effect of temperature on mass transfer and thermodynamic parameters in the removal of amoxicillin via hollow fiber supported liquid membrane

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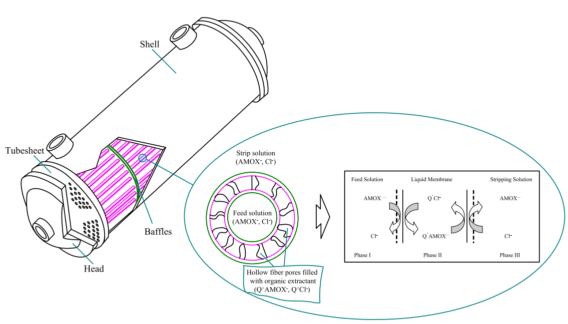
HIGHLIGHTS

- The effect of temperature on the separation of amoxicillin via HFSLM was studied.
- The highest percentage of extraction of amoxicillin was 89.65% at 318.15 K.
- The activation energy (E_a) of amoxicillin extraction was found to be 44.28 kJ/mol.
- Amoxicillin extraction was endothermic $(-\Delta H^0_{ex})$ while $(+\Delta S^0_{ex})$ and $(-\Delta G^0_{ex})$ were forward reaction.
- $k_m < k_f$, implying that mass transfer in membrane phase was rate controlling step.

GRAPHICAL ABSTRACT



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ABSTRACT

The influence of temperature on the separation of amoxicillin via HFSLM containing the carrier Aliquat 336 was systematically investigated. Mass-transfer parameters including distribution ratio and flux as well as thermodynamic properties namely ΔH and ΔG were also determined at different temperatures ranging from 278.15 K to 318.15 K. The positive value of the enthalpy change (24.5778 kJ/mol) indicated that the extraction process is endothermic reaction. Further, the positive value of the entropy change (112.0145 J/mol K) and the negative value of the Gibbs free-energy indicated that the extraction process is forward reaction. It was found that by increasing the temperature of the system from 278.15 to 318.15 K, extraction of amoxicillin increased from 81.81% to 89.65% and the stripping of amoxicillin increased from 76.63% to 84.70% respectively. Activation energy (E_a) of amoxicillin extraction was found to be 44.28 kJ/mol. This implied that the chemical reaction was the mass transfer controlling step. The mass transfer coefficients in the membrane phase were found to be less than the mass transfer coefficients in the feed phase within the range of temperatures i.e. 278.15–318.15 K. This implied that the mass transfer process in the membrane phase was the rate controlling step.

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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 (–)
D	distribution ratio of amoxicillin (–)
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	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	volumetric flow rate (cm^3/s)
L	length of the hollow fiber (cm)
N	number of hollow fibers in the 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)
T	Temperature (K)

Greek letters

τ	tortuosity of membrane (–)
ε	porosity of membrane (–)

Subscripts

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

1. Introduction

Amoxicillin ($C_{16}H_{19}N_3O_5S$) is an antibiotic which is widely used to treat various infections in both human beings and animals [1,2]. However, when amoxicillin is released into the environment, it is a major problem. When amoxicillin, for example, is introduced into surface water and groundwater, a change in aquatic ecosystems takes place [3,4]. This is owing to insufficient removal of amoxicillin in the conventional water and wastewater treatment plant. Thus, bacteria develop causing resistance to these drugs in humans and further failure of treatment with antibiotics [5–7]. Many applications for antibiotic removal from wastewater are available including ion exchange, adsorption, ultraviolet radiation, chemical coagulation and flocculation, ozonation, chlorination, membrane reverse osmosis and ultra filtration, and biological degradation [8–10].

Membrane technology has attracted much research for industrial wastewater treatment [11–13]. Recently, hollow fiber supported liquid membrane (HFSLM) has emerged as the technology of interest for chemical and environmental engineers in the treatment of wastewater containing low concentrations of metal ions, pharmaceuticals and organic compounds [14–16]. The principle of HFSLM is based on the simultaneous extraction and stripping of contaminant compounds with appropriate solvents in a single stage operation [17]. The advantages of such a system over other techniques include high treatment efficiency and selectivity, low energy costs, less solvent used and it is a simple design amenable to scaling up for industrial applications [18–22].

It is clear therefore that the HFSLM system should be suitable for the treatment of wastewater containing amoxicillin (see Fig. 1). Based on the literature, temperature was cited as one of the important operating parameters since it affected the performance of the mass transfer process during extraction and stripping [23,24]. Moreover, the effect of temperature is of significance to chemical reactions as denoted by the Arrhenius equation. Yet, few researchers have reported on the effect of temperature and thermodynamic parameters i.e. ΔG and ΔH on the mass transfer process in HFSLM [25,26]. Thus, the objective of this work is to investigate the effect of temperature on the mass transfer process during the extraction and stripping of amoxicillin via HFSLM. The work also employs Van't Hoff Model analysis of selectivity values derived from variable temperatures in order to assess the thermodynamic functions of separation.

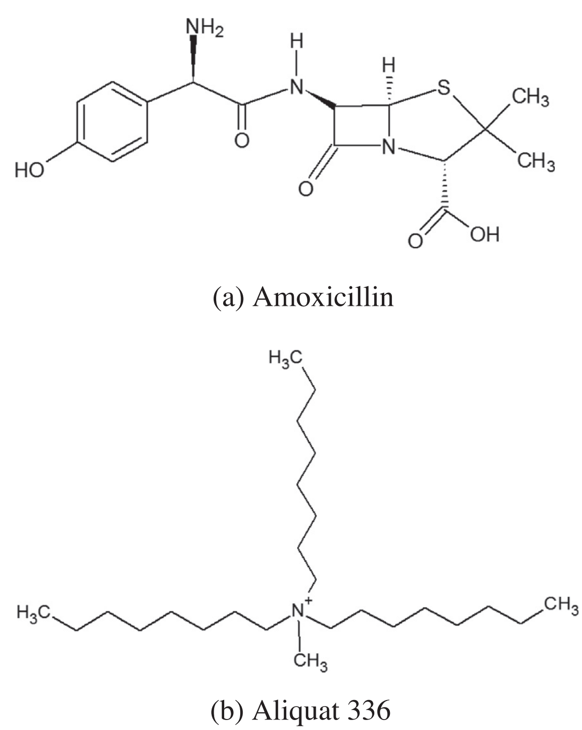

Fig. 1. Chemical structures: (a) amoxicillin and (b) Aliquat 336.

2. Theory

The extraction and recovery of the botanical extract amoxicillin (AMOX) [27,28] and Aliquat 336 (QCl) is shown in Eqs. (1) and (2), respectively (see Fig. 1). Fig. 2 shows the mechanism and transport kinetics scheme of amoxicillin through the hollow fiber supported liquid membrane. As shown in Eq. (1), amoxicillin in anionic form reacts with the extractant (i.e. QCl) to form complexes which will be recovered according to Eq. (2) with NaCl as stripping solution. Fig. 2 illustrates the use of HFSLM to separate amoxicillin from feed solution:

$$AMOX^-_{(f)} + Q^+Cl^-_{(m)} \underset{k_2}{\overset{k_1}{\leftrightarrow}} Q^+AMOX^-_{(m)} + Cl^-_{(f)} \quad (1)$$