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Kinetic Studies of Copper(II) Extraction from EDTA-Bearing Solutions by Aliquat 336 Using a Membrane Cell

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ABSTRACT

The kinetics of extraction of copper(II) from aqueous solutions containing an equimolar amount of EDTA (ethylenediaminetetraacetic acid) with Aliquat 336 (a quaternary amine) in kerosene were studied using a microporous membrane-based stirred cell. This device could give the intrinsic reaction rate for the formation of the amine complex occurring at or near the organic–aqueous interface by eliminating the diffusion resistances. Experiments were performed as a function of aqueous pH (3–11), copper(II) concentration (0.79–3.14 $\text{mol}\cdot\text{m}^{-3}$), Aliquat 336 concentration (50–500 $\text{mol}\cdot\text{m}^{-3}$), and temperature (288–318 K). The experimental results showed that Aliquat 336 would adsorb at the interface and react with the Cu-EDTA chelated anions.

Key Words. Extraction kinetics; Copper(II); EDTA; Aliquat 336; Membrane-based stirred cell

INTRODUCTION

Discharges from metal-finishing industries and the manufacturing process of printed circuit boards for electrodeless copper plating (1, 2), and washing effluents of remediation of metal-contaminated soils (3, 4) often contain metal ions and complexing agents. The presence of strong complexing agents such as EDTA (ethylenediaminetetraacetic acid), NTA (nitrilotri-

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acetic acid), citrate, and tartarate may make the chemical precipitation methods involving hydroxides and sulfides less effective, even with relatively high levels of metals (5). Furthermore, the high buffer capacity provided by the complexing agents requires excessive amounts of chemicals to neutralize the alkalinity. These problems and restrictions warrant investigations on other recovery alternatives.

In general, the recovery methods depend greatly on the particular complexing agents and metals involved as well as on their concentrations. Both ion exchange and reverse osmosis have high operation and maintenance costs for dilute solutions and are subject to fouling. The adsorption performance of activated carbon was found to be unsatisfactory although it is widely used for the treatment of organic pollutants (2). Our recent studies showed that Cu(II) can be effectively coextracted with EDTA in the form of chelated anions from aqueous solutions using Aliquat 336 as long as the pH is not far beyond 7 (6). The high distribution ratio provides the possibility of using liquid–liquid extraction for removal of Cu(II) from such chelated solutions.

In this paper the kinetics of such an extraction system are studied. These data would be useful in process design of practical applications including the so-called nondispersive solvent extraction using hollow fiber modules. In fact, Alonso and his coworkers (7–11) modeled the mass transfer of CrO_4^{2-} with Aliquat 336 in hollow fiber extractors. Due to the lack of a kinetic scheme of extraction reactions, they always assumed that the reactions occurring at the interface are fast compared to diffusion in the membrane, organic layer, or aqueous layer.

It is known that a Lewis-type transfer cell cannot be successfully applied to obtain the “intrinsic” kinetics of chemical reactions taking place at or near the organic–aqueous interface because the mass-transfer effect cannot be correctly quantified (12). A simple and easy-to-operate membrane-based stirred cell, resembling a rotating diffusion cell (13), has been used to examine the interfacial kinetics of metal extraction with organophosphoric acids (14). This device allows the contribution of diffusion resistances to the overall process to be estimated and thus the intrinsic kinetics of the interfacial reactions to be obtained. In this work the interfacial reaction rates of copper(II) extraction from EDTA-bearing streams with Aliquat 336 in kerosene are measured. The effect of temperature on the reaction rates is also investigated.

EXPERIMENTAL

Apparatus, Membrane, and Reagents

The membrane-based stirred cell used was the same as that described previously but with slightly larger dimensions (14). Two chambers separated by a membrane support of 44.2-cm^2 area were stirred at the same rate (400 rpm) but in opposite directions. The microporous PVDF (polyvinylidiene fluoride) hydrophobic membrane used (GVHP, Millipore) had a thickness of 125 μm, an average pore size of 0.22 μm, and a typical porosity of 75%.

Aliquat 336 (tri-*n*-octylmethylammonium chloride, FW 404.16) was supplied by Aldrich Co. Prior to use it was washed with dilute HCl solution to remove water-soluble impurities. Kerosene, from Union Chemical Works Ltd., Taiwan, was washed twice with 20 vol% H_2SO_4 to completely remove aromatics before use. It was finally washed with deionized water (Millipore Milli-Q) three times. EDTA, $CuSO_4$, and other inorganic chemicals were supplied by Merck Co. as analytical reagent grade.

The organic phase was prepared by diluting Aliquat 336 in kerosene, and the initial amine concentration ranged from 50 to 500 $mol \cdot m^{-3}$. The aqueous phase was prepared by dissolving equimolar amounts of $CuSO_4$ and EDTA in deionized water, for which the pH was adjusted by adding a small amount of HCl or NaOH. The initial concentration of Cu(II) ranged from 0.79 to 3.14 $mol \cdot m^{-3}$ and the initial pH varied from 3 to 11.

Experimental Procedures

Time profiles of aqueous-phase concentrations of Cu(II) were measured as follows. The organic phase (320 cm^3) was first placed in the lower chamber. The membrane, presoaked with the organic phase, was clamped, and the cell was assembled. An equal volume of the aqueous phase was then poured into the upper chamber. Timing was started upon addition of the aqueous phase. The temperature was controlled in the 288–318 K range. Aqueous samples (5 cm^3) were taken at preset time intervals, and the concentration of Cu(II) was analyzed by a GBC atomic absorption spectrophotometer (Model 932). The concentration of Cu(II) in the organic phase was obtained by mass balance. The aqueous pH was measured using a pH meter (Horiba Model F-23). Each experiment was at least duplicated under identical conditions.

RESULTS AND DISCUSSION

Determination of Mass Transfer Coefficients

EDTA, a polyprotic acid (H_4L), exists in a number of protonated forms in aqueous solutions. It readily forms stable complexes with most heavy metals in a 1:1 molar ratio. In principle, the distribution of species can be obtained from mass-balance equations of metals and EDTA. For a diluted equimolar (10 $mol \cdot m^{-3}$) solution of Cu(II) and EDTA, it was shown that the divalent chelated anions CuL^{2-} absolutely predominate at pH 3–12 and the univalent chelated anions $CuHL^-$ at pH < 3 (15). These information enabled us to know the extractable aqueous species at different pH values.

TABLE 1
Transport Parameters Evaluated in This Membrane-Based Stirred Cell at 298 K

System (solute/medium)	Diffusivity ($m^2 \cdot s^{-1}$)	Mass-transfer coefficient ($m \cdot s^{-1}$)
Iodine/water	1.39×10^{-9}	$k_a = 7.20 \times 10^{-5}$
Iodine/kerosene	2.76×10^{-9}	$k_o = 9.02 \times 10^{-5}$
Iodine/membrane[a]		$k_m = 7.60 \times 10^{-6}$
CuL^{2-}/water	4.71×10^{-10}	$k_a = 3.64 \times 10^{-5}$
Aliquat 336/kerosene	2.35×10^{-9}	$k_o = 8.13 \times 10^{-5}$
Aliquat 336/membrane[a]		$k_m = 6.41 \times 10^{-6}$
Aliquat 336 complex/kerosene	2.14×10^{-9}	$k_o = 2.58 \times 10^{-5}$
Aliquat 336 complex/membrane[a]		$k_m = 5.84 \times 10^{-6}$

[a] Pores of the membrane were fully impregnated with kerosene.

For immiscible organic and aqueous phases separated by a hydrophobic microporous membrane, the individual mass transfer coefficients in the aqueous and organic stagnant layers, and within the membrane (completely wetted by the organic phase), k_a, k_o, and k_m, respectively, can be determined from flux data of more hydrophilic (e.g., acetic acid) and hydrophobic solutes (e.g., iodine) (14). Owing to the ease of concentration analysis, the mass transfer coefficient k_m of the complex, rather than amine itself, was experimentally verified in this work. The flux was obtained using the same cell, where the complex was transferred from a preloaded organic phase (complex concentration, 40 $mol \cdot m^{-3}$) across the same membrane to a fresh organic phase. The concentration of the complex was analyzed at preset time intervals by an atomic absorption spectrophotometer. Due to the lack of an organic–aqueous interface, k_m was obtained from time profiles of the complex concentration which equals $1/[(1/k_m) + (2/k_o)]$. The individual mass transfer coefficient of the complex in the organic layer k_o is estimated as below.

Here, the values of k_a for the Cu(II) chelated anions, k_o and k_m for the amine, and k_o for the complex are estimated from k_a and k_o for iodine and k_m for the complex based on the relationships that $k_m \propto D_j$ and k_a (or k_o) $\propto D_j^{2/3} \nu^{-1/6}$, where ν is the kinematic viscosity of the media (16). The results are listed in Table 1. In this correlation the diffusivities of species D_j are estimated by the Hayduk and Minhas equation (17). The value of ν for kerosene is 7.67×10^{-7} $m^2 \cdot s^{-1}$ at 298 K. The molar volume of kerosene at the normal boiling point was found to be 278 $cm^3 \cdot mol^{-1}$ by the Tyn and Calus method (17). The parachors for kerosene, Aliquat 336, and the Aliquat 336 complex are estimated to be about 510, 1079, and 1609 $cm^3 \cdot g^{1/4} \cdot s^{-1/2}$, respectively, by the method of additive group contributions (17). It is noted that the complex contains two molecules of the amine per molecule of the chelated anions in the ranges studied (6). Also, the values of k_m for amine and the complex taken here are assumed to be constant at all the extractant concentrations for simplicity.

Determination of Transfer (Extraction) Rates

It was reported that a hydrophobic microporous membrane with a high porosity and a pore size between 10^{-3} and 10^2 μm is not readily wetted by water (18). On the other hand, hydrocarbons and most other organic solvents readily wet it. Hence, the concentrations of species at the membrane–organic interface are considered to be identical because the interface is essentially homogeneous due to the high porosity of the membrane (19).

Typical time profiles of organic-phase concentrations of the chelated Cu(II) during extraction are shown in Fig. 1. The transfer rate J_f is obtained by the initial-rate technique according to

$$J_f = (V_o/S)(d[\overline{\mathrm{CuL}}]/dt)_{t=0} \qquad (1)$$

where V_o is the volume of the organic solution, S is the membrane cross-sectional area, and $[\overline{\mathrm{CuL}}]$ denotes the concentration of the amine complex in the organic phase.

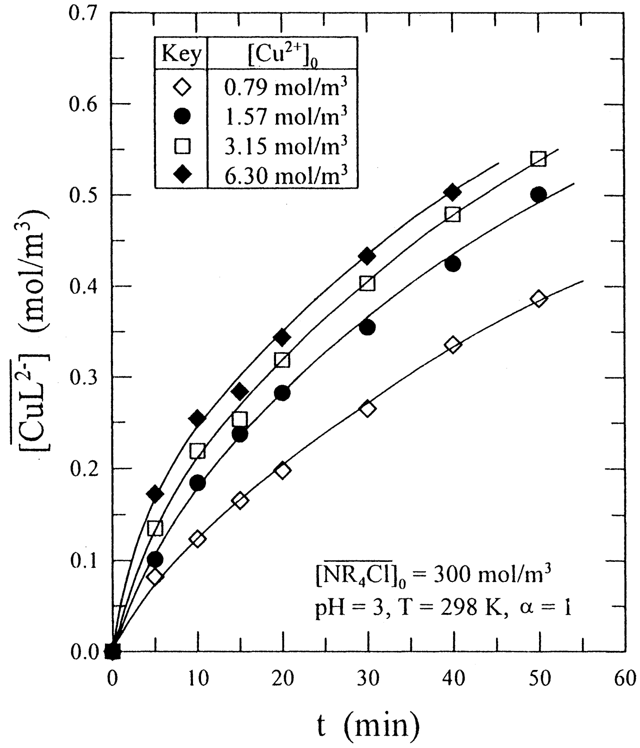

FIG. 1 Time profiles of organic-phase concentration of the amine complex at 298 K.

Intrinsic Interfacial Reaction Rates

The solubilities of Aliquat 336 and kerosene in water are only 2.03 and 0.19 dm^3/m^3, respectively, at 293 K (20). It is known that Aliquat 336 is a surface-active agent (21); i.e., it tends to accumulate near the interface of two immiscible phases. Reaction between the chelated anions and amine is consequently considered to occur at the interface on the aqueous side to form the complex.

In a real extraction process the overall process is determined not only by interfacial chemical reactions but also by diffusion of species toward and away from the interface (12). In order to eliminate the contribution of diffusion resistances to the overall process, an attempt was made to estimate the concentrations of species at or near the interface and to correlate them with rate data. For a pseudosteady-state approximation, the concentration of species adjacent to the interface is obtained from the measured transfer rate J_f, in which the interfacial reaction rate equals the mass transfer rate (14). This type of approximation is commonly used in most membrane operations. The membrane itself contains very little solution (0.05 cm^3) compared to both adjacent chambers. Hence, the profile across the membrane is always close to its steady value, even though the chamber concentrations are time-dependent (22).

For the extraction of chelated anions with Aliquat 336 in kerosene, we thus have

$$J_f = R_f - R_b = k_{CuL}([CuL^{2-}]_b - [CuL^{2-}]_i) = K_{\overline{NR_4Cl}}([\overline{NR_4Cl}]_b - [\overline{NR_4Cl}]_i) \quad (2)$$

where the subscripts "i" and "b" refer to the aqueous–membrane interface and bulk phase, respectively. R_f and R_b are the intrinsic reaction rates for the formation and dissociation of the amine complex, respectively. $K_{\overline{NR_4Cl}}$ is the overall mass transfer coefficients of Aliquat 336 across the membrane and organic layer, which equals $1/[(1/k_m) + (1/k_o)]$.

In this case the contribution of the reverse reaction is ignored; i.e., $R_f = J_f$, at least at the beginning of extraction. This is because initially the aqueous phase contains the chelated anions and the organic phase consists of free amine only. The reaction rates at different interfacial concentrations of the chelated anions are illustrated in Fig. 2, and they indicate a half order with respect to the chelated anions. Figure 3 shows the reaction rates at different interfacial concentrations of Aliquat 336. It is evident that the rate reveals a Langmuir-type dependence with respect to amine concentration when the pH is not more than 7 (which is checked by Fig. 4). As indicated in an equilibrium study (6), at a sufficiently high amine concentration the distribution ratio first increases with increasing pH but then decreases as pH > 7. This is because the competitive reactions (Eq. 3) of OH^- and amine with the chelated anions oc-

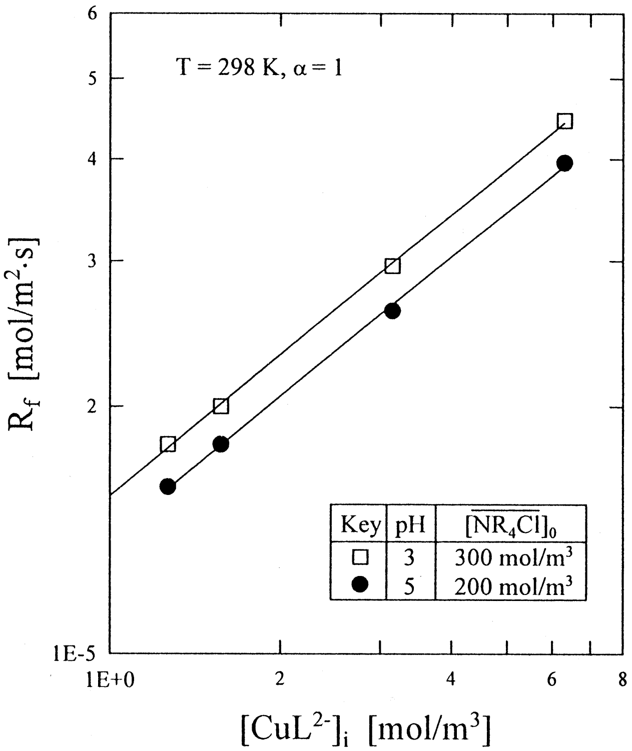

FIG. 2 Effect of interfacial concentration of the Cu(II) chelates on reaction rate at 298 K.

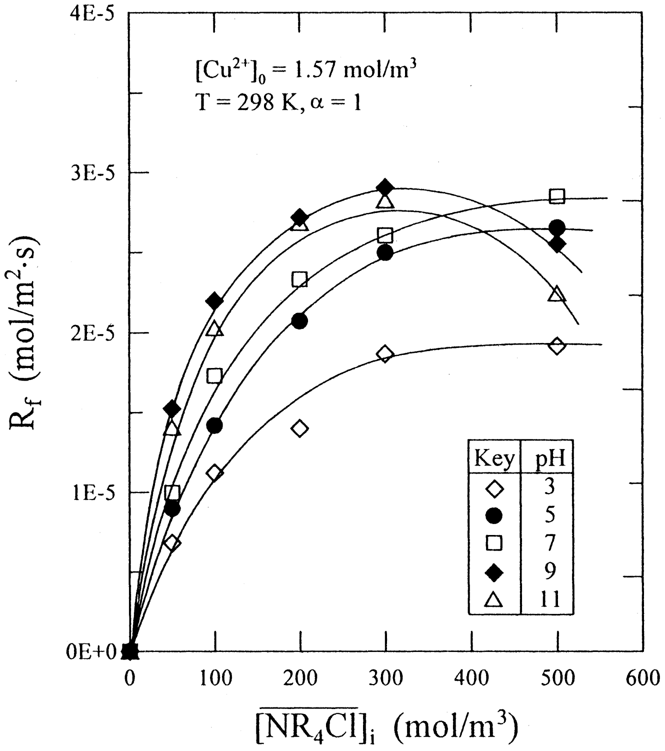

FIG. 3 Effect of interfacial concentration of Aliquat 336 on reaction rate at 298 K.

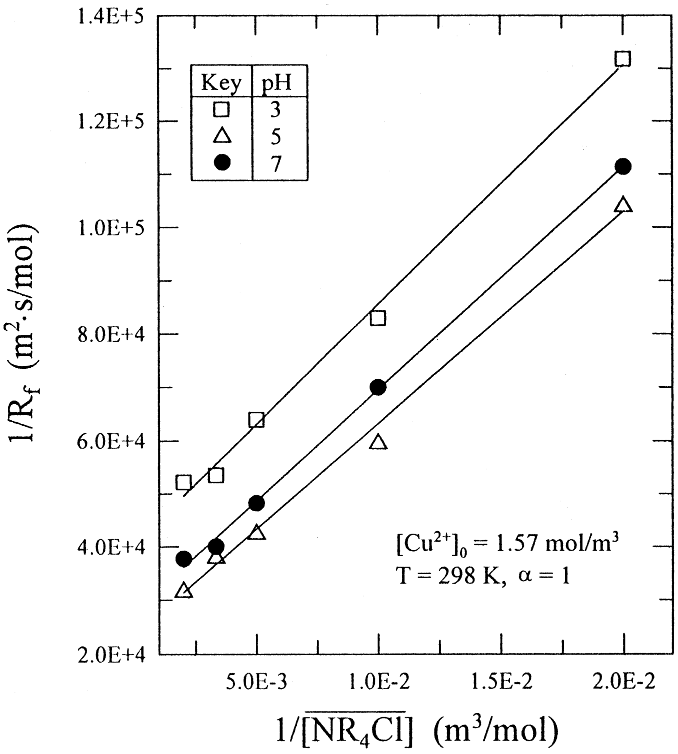

FIG. 4 The Langmuir plot of reaction rate and interfacial concentration of Aliquat 336 at 298 K.

cur to some extent (6) when the pH is greater than 7.

$$OH^- + \overline{NR_4Cl} \Leftrightarrow \overline{NR_4OH} + Cl^- \tag{3}$$

As indicated above, the univalent anions $CuHL^-$ dominates in the aqueous phase at $pH < 3$ and the extent of reaction Eq. (3) is much greater at $pH > 7$; both make the reaction stoichiometries complicated. Only the data obtained at pH 3–7 are henceforth considered. The reaction between the chelated anion CuL^{2-} and Aliquat 336 in kerosene can be given as follows (6):

$$CuL^{2-} + 2\overline{NR_4Cl} \Leftrightarrow \overline{(NR_4)_2CuL} + 2Cl^- \tag{4}$$

According to the results of Figs. 2 and 3, the reaction rate for complex formation of Aliquat 336 and the chelated anions at pH 3–7 is thus expressed as

$$R_f = \frac{k_1[CuL^{2-}]^{1/2}[\overline{NR_4Cl}]}{1 + k_2[\overline{NR_4Cl}]} \tag{5}$$

The rate constants k_1 and k_2 in Eq. (5) are found to be 1.97×10^{-7} $m^{5/2}\cdot mol^{-1/2}\cdot s^{-1}$ and 7.37×10^{-3} $m^3\cdot mol^{-1}$, respectively, at pH 3 and 298 K.

From the rate data and Eq. (2), the maximum differences between the interfacial and bulk concentrations of the chelated anions and Aliquat 336 are about 1.22 and 9.78 mol·m^{-3}, respectively, under the ranges studied (chelate concentration < 6.29 mol·m^{-3}, amine concentration < 500 mol·m^{-3}). The contributions of the diffusion of amine and the complex in the membrane and organic layer to the overall extraction process play a comparatively more important role.

Figure 5 shows the reaction rates over an extended pH range of 3–11. The pH effect appears to be negligible although the rate does increase with increasing pH. It is expected that the side reaction of Eq. (3) occurs to a large extent at high pH; however, such an effect is significant only in the case of high amine concentration. By substituting the data of Fig. 5 into Eq. (5), we can obtain the pH trends of k_1 and k_2 (Fig. 6). It is evident that increasing pH leads to a slight increase in k_1 and a slight decrease in k_2, which will be explained in the following section.

Comments on the Rate Equation

The activation energies of the reaction between the chelated anions and Aliquat 336 at pH 3 are found to be 52.8 and 34.1 kJ·mol^{-1} from Arrhenius plots

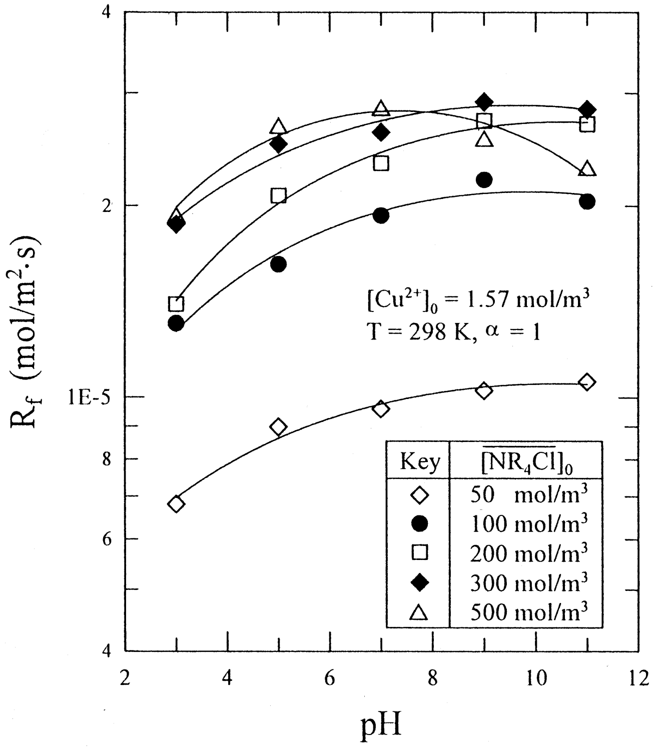

FIG. 5 Effect of aqueous pH on the reaction rate at 298 K.

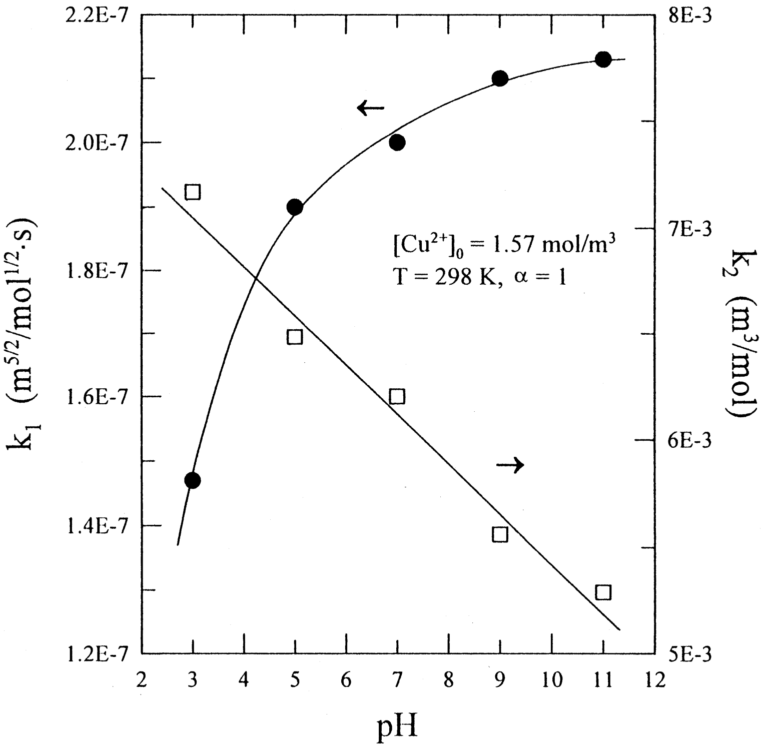

FIG. 6 Effect of aqueous pH on the reaction rate constants at 298 K.

of the rate constants k_1 and k_2, respectively (Fig. 7). The formation of hydrophobic complexes in the liquid–liquid extraction processes generally has an activation energy greater than 40–80 kJ·mol^{-1} (23). The slightly low activation energy for k_2 obtained here will be explained below. An attempt was made to present a reaction mechanism based on the equilibrium and kinetic results. However, this attempt failed because of the complicated, nonideal behavior of the organic-phase formulation (6).

It was well known that the extractant Aliquat 336 is a highly surface-active agent and tends to adsorb at the organic–aqueous interface (21). This can explain the reason why Eq. (5) has a form similar to those found for heterogeneously catalyzed reactions with a Langmuir-type adsorption isotherm (24). As shown in Fig. 5, the rate constant k_2 is less sensitive to temperature than k_1. This confirms that k_2 is related to the adsorption constant because the adsorption/desorption at a liquid–liquid interface has a lower energy change than that of the chemical reaction itself. In addition, increasing the pH results in the conversion of the chloride salt of the amine to the hydroxide salt. This decreases the interfacial activity (21).

Finally, an explanation for the half order with respect to concentration of the chelated anions (Eq. 5) cannot be given at this stage. However, it does re-

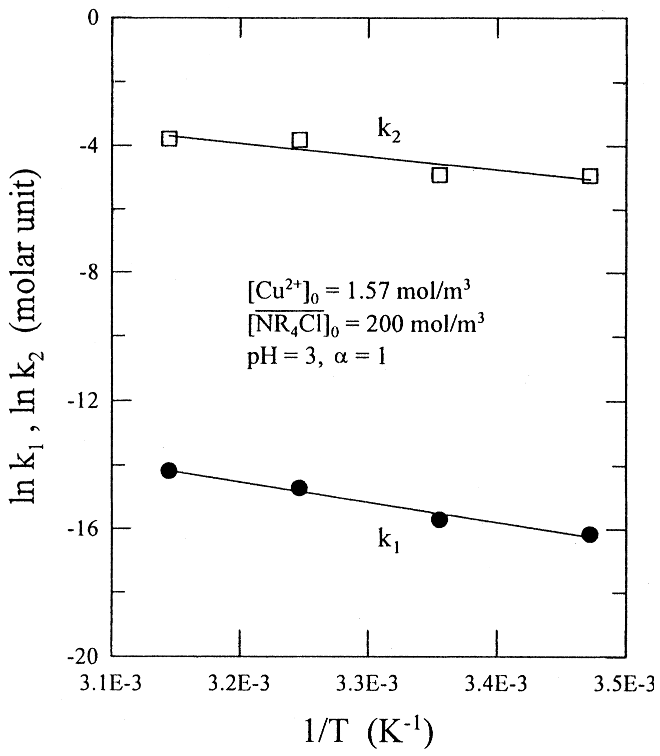

FIG. 7 Temperature dependence of the reaction rate constants of complex formation.

semble the adsorption of diatomic gases with dissociation; i.e., one molecule of the anion reacts with two molecules of the amines preadsorbed at the surface sites to form complexes which are weakly adsorbed.

CONCLUSIONS

Interfacial transfer kinetics for the extraction of EDTA-chelated Cu(II) anions (CuL^{2-}) from aqueous solutions with Aliquat 336 ($\mathrm{NR_4Cl}$) in kerosene have been studied using a membrane-based stirred cell in the 288–318 K temperature range. Rate data are correlated with the interfacial concentrations of reactive species, which allows the intrinsic kinetics of interfacial reactions to be obtained. The reaction rates (R_f) for formation of the amine complex can be expressed as $R_f = k_1[\mathrm{CuL}^{2-}]^{1/2}[\overline{\mathrm{NR_4Cl}}]/(1 + k_2[\overline{\mathrm{NR_4Cl}}])$. This equation implies that Aliquat 336 tends to adsorb at the interface, which is supported by the fact that k_2 is less sensitive to temperature than k_1. The activation energies for k_1 and k_2 at pH 3 are found to be 52.8 and 34.1 kJ·mol^{-1}, respectively. A half order with respect to the chelated anions probably relates to the stoichiometry; i.e., one molecule of the anion reacts with two molecules of the amines preadsorbed at the surface sites.

NOMENCLATURE

Aliquat 336	tri-*n*-octylmethylammonium chloride (NR_4Cl)
D_j	diffusivity of species j in bulk liquid phase ($m^2 \cdot s^{-1}$)
EDTA	ethylenediaminetetraacetic acid (H_4L)
J	transfer rate of the chelated anions ($mol \cdot m^{-2} \cdot s^{-1}$)
K	overall mass-transfer coefficient ($m \cdot s^{-1}$)
k	individual mass-transfer coefficient ($m \cdot s^{-1}$)
k_1, k_2	rate constants for formation of the complex defined in Eq. (5)
R_b	intrinsic reaction rate of dissociation of the complex ($mol \cdot m^{-2} \cdot s^{-1}$)
R_f	intrinsic reaction rate of formation of the complex ($mol \cdot m^{-2} \cdot s^{-1}$)
S	membrane cross-sectional area (m^2)
t	time (s)
T	absolute temperature (K)
V	volume (m^3)
[]	molar concentration of species in the brackets ($mol \cdot m^{-3}$)

Greek Letters

α	initial concentration ratio of EDTA to Cu(II) in the aqueous phase
ν	kinematic viscosity of the medium ($m^2 \cdot s^{-1}$)

Subscripts

a, m, o	aqueous, membrane, and organic phases, respectively.
b	bulk
i	aqueous–membrane interface
0	initial

Superscript

$\overline{\text{(overbar)}}$	species in the organic (membrane) phase

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