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## Facilitated Transport of Gold from Cyanide or Chloride Media by Cyanex 923

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### ABSTRACT

The commercially available extractant Cyanex 923 (phosphine oxides mixture) was studied to be applied in the carrier-facilitated transport of gold, from either cyanide or chloride aqueous media, across a polymer-immobilized liquid membrane (PILM).  $\text{Au}(\text{CN})_2^-$  is transported from alkaline pH values. The presence of lithium salts (and the respective counter anion) in the media improves the transport, whereas the organic diluent seems not to influence the metal permeability. Water can be used effectively, as receiving phase, for the present system. In chloride media, the carrier is able to transport gold(III), increasing the permeability as the initial HCl concentration is increased (up to  $5 \times 10^{-1}$  M HCl). This permeability is dependent on the respective organic diluent, being the metal flux independent at higher initial gold concentrations but metal dependent at lower gold concentrations in the feed phase. Sodium thiocyanate solutions can be used effectively as receiving phase in this

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media. From the experimental data, the mass-transfer coefficient in the aqueous feed phase and the thickness of the aqueous boundary layer were calculated for both, cyanide and chloride, aqueous media.

*Key Words:* Facilitated membrane transport; Gold; Cyanex 923.

## INTRODUCTION

Recovery of gold from various resources is restricted to leaching in cyanide or aqua regia media. From the leaching solutions, various procedures have developed for the separation and obtention of this precious metal.<sup>[1,2]</sup> From separation technologies, liquid membranes (LMs) have been emerging as an attractive alternative to solvent extraction, having different applications in the field of separation sciences,<sup>[3,4]</sup> with potential uses that cover the separation of hydrocarbons, metals, inorganic and organic species, and biotechnology, and medical applications. When the transport process takes place in the presence of an extractant contained within the membrane, facilitated transport occurs, which can be used as a technique for the selective separation of two or more solutes containing in the same feed phase. This is without a doubt an advantage over other less-selective separation techniques. On the other hand, lack of membrane stability is the major drawback associated with the use of supported liquid membranes; the instability of membranes may be due to different causes, i.e., loss of carrier phase by its solubility in both aqueous phases, the osmotic pressure gradient across the membrane, membrane pore diameter, etc.<sup>[5–7]</sup> Various approaches can be found in research that seek to determine supported liquid membrane stability; whereas, several avenues proposed for improvement of membrane stability include synthesis of selective carriers that possess very low solubility in water, modifications of the aqueous phase to achieve high organic/water interfacial tension, different degrees of binding the carrier phase to the polymeric membrane, and utilization of alternative configuration systems.<sup>[7]</sup>

Some drawbacks encountered in the traditional gold separation processing had led, during the past years, to investigation of new extraction processes based on liquid membranes for the recovery of gold from various aqueous solutions.<sup>[8–14]</sup>

Among the various carriers used for gold transport in supported liquid membrane systems (SLMs), there is no apparent information about the use of phosphine oxides as carriers in the polymer-immobilized liquid membrane (PILM) configuration for gold transport in cyanide or chloride media.

In the present study, the transport of gold from these two aqueous media through a PILM using the phosphine oxide Cyanex 923 as carrier was studied.

## EXPERIMENTAL

The composition of the phosphine oxide Cyanex 923 is based on up seventeen alkyl-phosphine oxides,<sup>[15]</sup> with an average molecular weight of 348 and a density at 20°C of 880 kg/m<sup>3</sup>. Cyanex 923 was obtained from CYTEC Ind. (Niagra Falls, Ontario, Canada). Table 1 shows the main characteristics of the organic diluents used in the present investigation. These, and all the chemicals used, were AR grade. Gold cyanide solutions were prepared by dissolving KAu(CN)<sub>2</sub> in distilled water. The different concentrations used in the experiments were prepared from a standard solution of  $2.5 \times 10^{-3}$  M in gold(I). The pH of the solution was adjusted by small additions of diluted sodium hydroxide solutions. During the test, the pH was continuously controlled using a Crison 605 pH meter (Alella, Barcelona, Spain). In chloride media, the gold(III) solutions were prepared in a similar manner as HAuCl<sub>4</sub>.

The transport experiments were carried out using the same cell, membrane support, and procedure as described in a previous work.<sup>[16]</sup> The characteristics of the support were:  $12.5 \times 10^{-3}$  cm, thick microporous polyvinylidenedifluoride film, 75% nominal porosity, effective pore size of  $2.2 \times 10^{-5}$  cm, and tortuosity 1.67. The effective membrane area was 11.3 cm<sup>2</sup>.

Gold concentration in the feed phases was analyzed by atomic absorption spectrometry, using a Perkin Elmer model 1100B (Madrid, Spain) spectrophotometer, though in several experiments, the gold concentration in the receiving phases was also analyzed to verify active metal transport. The gold concentration in the aqueous solution was found to be reproducible within  $\pm 2\%$ . Transport experiments ran for 3 hours and the permeation coefficient (P) was computed using Eq. (1):

$$\ln \frac{[Au]_t}{[Au]_0} = -\frac{A}{V}Pt \quad (1)$$

**Table 1.** Characteristics of the diluents used in gold transport with Cyanex 923.

Diluent	Boiling range (°C)	Aromatics (%)	Density (kg/m <sup>3</sup> )	Viscosity (cP)
Xylene <sup>a</sup>	139–142	100	863	0.6
<i>n</i> -Decane	168–178	—	730	0.9
Cumene	153	100	862	0.7
Toluene	110	100	867	0.6

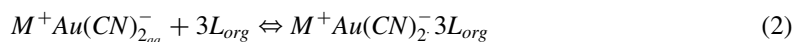
<sup>a</sup>Mixture of ortho-meta and para-xylene.

where  $V$  is the volume of the feed phase solution,  $A$  is the effective membrane area,  $[Au]_0$  and  $[Au]_t$  are the concentrations of metal ion in the feed phase at time zero and at given time, and  $t$  is the elapsed time. Additional experiments were performed to study the membrane stability, results obtained show that the permeability values are constant and reproducible within  $\pm 2\%$ .

## RESULTS AND DISCUSSION

### Gold Transport from Cyanide Media

In the pH range of 6 to 11, the extraction of gold(I) cyanide by Cyanex 923 is represented by the general equilibrium,<sup>[17]</sup>



which is pH-independent and where *aq* and *org* denote species in the aqueous and organic phases, respectively.  $L$  represents the active substance of the extractant. In this reaction,  $M^+$  represents a cation, such as sodium, potassium, or lithium (preferably).

### Influence of Stirring Speed of the Feed and Receiving Phases

The influence of the stirring speed in both phases on the permeability of gold(I) ( $2.5 \times 10^{-5}$  M) with Cyanex 923 ( $7.5 \times 10^{-1}$  M in xylene) is shown in Table 2. As the stirring speed of the feed phase is increased to  $1400 \text{ min}^{-1}$ , the thickness of the aqueous boundary layer near to the membrane feed interface

**Table 2.** Influence of the stirring speed (feed phase) on gold permeation.

Stirring speed ( $\text{min}^{-1}$ )	P (cm/s)
1000	$2.7 \times 10^{-3}$
1200	$3.2 \times 10^{-3}$
1400	$3.4 \times 10^{-3}$
1600	$3.4 \times 10^{-3}$

Feed phase: 1 M LiCl at pH  $10.0 \pm 0.1$ .  
Membrane support: Durapore GVHP 4700.  
Receiving phase: water.

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is minimized to a constant value, which resulted in the highest permeability. On the other hand, the variation of the stirring speed in the receiving phase has a negligible effect on the variation of gold permeation. Therefore, stirring speeds of  $1400 \text{ min}^{-1}$  and  $1000 \text{ min}^{-1}$  were selected in the feed and receiving phases, respectively, to avoid concentration polarization conditions in the aqueous feed phase.

**Influence of Receiving Phase Composition**

The variation in the composition of the receiving phase (water or  $1 \times 10^{-1} \text{ M NaOH}$ ) on gold(I) permeation was studied using the same feed and organic solutions (on a Durapore GVHP4700 support) as described previously. Results obtained showed that this variation in the composition has a negligible effect on gold permeation ( $3.4 \times 10^{-3} \text{ cm/s}$  with water,  $3.6 \times 10^{-3} \text{ cm/s}$  with NaOH). Therefore, water was selected throughout the investigation.

**Effect of Ionic Strength**

Table 3 shows the results obtained from the transport of  $\text{Au}(\text{CN})_2^-$  in the presence of different salts. The organic phase used in the experiments contained Cyanex 923 ( $7.5 \times 10^{-1} \text{ M}$ ) in xylene. The aqueous phase contained

**Table 3.** Effect of the presence of different salts on  $\text{Au}(\text{CN})_2^-$  transport.

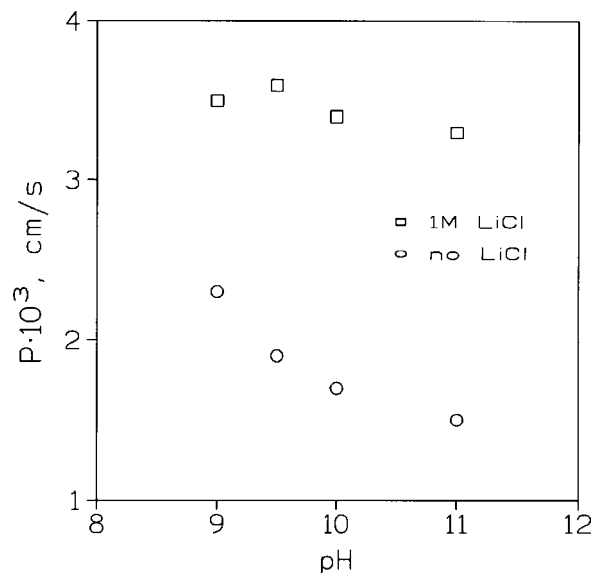
Salt	P (cm/s)
—	$1.7 \times 10^{-3}$
$\text{LiNO}_3$	$4.8 \times 10^{-3}$
$\text{LiCl}$	$3.4 \times 10^{-3}$
$\text{Li}_2\text{SO}_4$	$2.0 \times 10^{-3}$
$\text{KNO}_3$	$1.5 \times 10^{-3}$
$\text{KCl}$	$1.1 \times 10^{-3}$
$\text{NaNO}_3$	$1.1 \times 10^{-3}$
$\text{NaCl}$	$8.8 \times 10^{-5}$

Feed phase: salts at 1 M. Membrane support: Durapore GVHP4700. Receiving phase: water.

$2.5 \times 10^{-5}$  M of gold(I) at  $\text{pH } 10.0 \pm 0.1$ . From the results, it can be observed that salts containing either  $\text{Na}^+$  or  $\text{K}^+$  cations have a relative influence on gold transport. However, the presence of the  $\text{Li}^+$  cations produces an increase of gold permeation. On the other hand, the presence of the counter anion has also an effect on gold permeation in the apparent order: nitrate > chloride > sulfate; however, by using NaCl, no practical gold transport is achieved.

To study the influence of the presence of lithium salts in the feed phase on gold permeation in more detail, a set of experiments was carried out at a constant LiCl concentration of 1 M and varying pH values.

In Fig. 1, the variation of  $P_{\text{Au}}$  is plotted vs. aqueous pH for the transport of  $\text{Au}(\text{CN})_2^-$  from aqueous solutions containing  $2.5 \times 10^{-5}$  M gold. Organic phases contained  $7.5 \times 10^{-1}$  M Cyanex 923 in xylene. It can be noted that in the absence of LiCl, the transport of gold(I) tends to decrease as the pH increases; whereas, in the presence of LiCl, the transport is higher at all pH values, and remains almost constant. This effect should be attributable to the fact that the transport occurs by solvation of ion pairs  $\text{Li}^+ \text{Au}(\text{CN})_2^-$  by the organic reagent as described in the gold-solvent extraction research using these phosphine oxides.<sup>[17,18]</sup>



**Figure 1.** Effect of the pH variation on gold(I) permeation using Cyanex 923 in xylene on a Durapore GVHP4700 support. Receiving phase: water.

### Organic Diluent Influence

Although at a first approximation, the diluent of the organic phase is just an inert component, in practice this is not always true. It should play an active part in the system, modifying the transport properties of several carrier system.<sup>[19–21]</sup>

Table 4, in which the permeation coefficients for gold transport are given, shows the effect of various diluents on gold(I) transport with Cyanex 923. The feed phase composition was  $2.5 \times 10^{-5}$  M Au(I) and 1 M LiCl at pH  $10.0 \pm 0.1$ . The organic phase contained  $7.5 \times 10^{-1}$  M Cyanex 923 in each diluent. As can be observed, in the present system, the change of the diluent has a negligible effect on  $\text{Au}(\text{CN})_2^-$  transport using Cyanex 923 as carrier.

### Effect of Extractant Concentration on Gold Transport

In Fig. 2 the gold permeability is plotted vs. the concentration of Cyanex 923 in the organic phase to study the effect of extractant concentration variation on gold(I) transport. Experiments were carried out at a constant pH value of  $10.0 \pm 0.1$ . Aqueous phases contained  $2.5 \times 10^{-5}$  M gold(I) and 1 M LiCl. In the organic phase, different extractant concentrations in xylene were used.

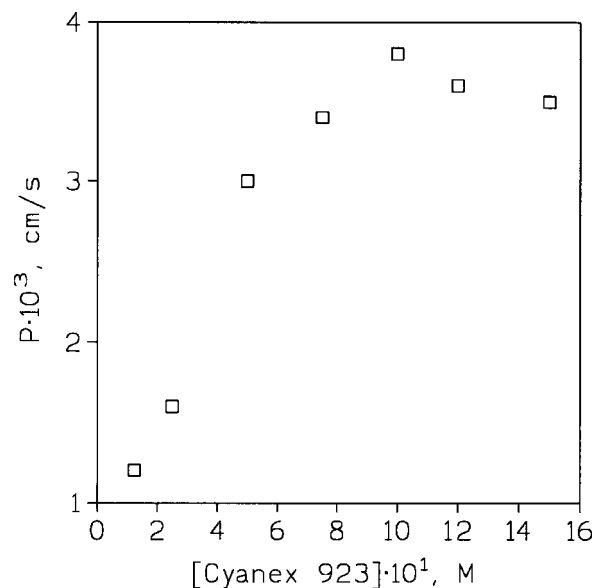
Results obtained revealed no significant change in permeability at higher carrier concentration. This constant permeability value, or limiting permeability ( $P_{\text{lim}}$ ), can be attributed to the assumption that diffusion in the membrane is negligible compared with the aqueous diffusion and the permeation process is controlled by the diffusion in the stagnant film of the feed phase. Thus,  $P_{\text{lim}} = 1/\Delta_{\text{aq}} = 3.6 \times 10^{-3}$  cm/s, and assuming a value of  $10^{-5}$  cm/s for  $D_{\text{aq}}$ ,<sup>[22]</sup> then  $d_{\text{aq}} = D_{\text{aq}}/P_{\text{lim}} = 2.8 \times 10^{-3}$  cm. This value is the minimum thickness of the stagnant aqueous diffusion layer in the present experimental conditions.

**Table 4.** Effect of organic phase diluent on gold permeability.

Diluent	P (cm/s)
Xylene	$3.4 \times 10^{-3}$
Cumene	$3.3 \times 10^{-3}$
<i>n</i> -Decane	$3.6 \times 10^{-3}$

Membrane support: Durapore  
GVHP4700. Receiving phase: water.





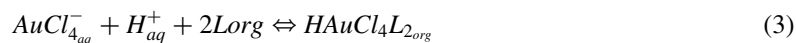
**Figure 2.**  $P_{Au}$  versus various Cyanex 923 concentrations. Membrane support: Durapore GVHP4700. Receiving phase: water.

### Effect of Initial Metal Concentration on Gold Transport

Table 5 shows the variation in the gold permeability and the initial flux ( $J = P[Au(I)]_0$ ) against the concentration of gold ranging from  $1.3 \times 10^{-5}$  to  $2.5 \times 10^{-4}$  M in the feed phase solution. It can be observed that with the present experimental conditions both the permeation coefficient and the metal flux increased with the increase of the initial gold concentration in the phase.

### Gold Transport from Chloride Media

The liquid–liquid extraction of Au(III) from HCl solutions by Cyanex 923 was also studied previously.<sup>[23]</sup> The extraction equilibrium is described by the following reaction:



where L is the organic extractant.

**Table 5.** Variation in the metal permeation and in the metal flux with the initial gold concentration.

[Au(I)] <sub>0</sub> (M)	P (cm/s)	J (mol/cm <sup>2</sup> s)
$1.3 \times 10^{-5}$	$2.2 \times 10^{-3}$	$2.8 \times 10^{-11}$
$2.5 \times 10^{-5}$	$3.4 \times 10^{-3}$	$8.6 \times 10^{-11}$
$1.3 \times 10^{-4}$	$5.7 \times 10^{-3}$	$7.2 \times 10^{-10}$
$2.5 \times 10^{-4}$	$6.3 \times 10^{-3}$	$1.7 \times 10^{-9}$

Feed phase: 1 M LiCl at pH  $10.0 \pm 0.1$ . Membrane phase:  $7.5 \times 10^{-1}$  M Cyanex 923 in xylene on a Durapore GVHP4700 support. Receiving phase: water.

### Influence of the Stirring Speed in the Feed Phase

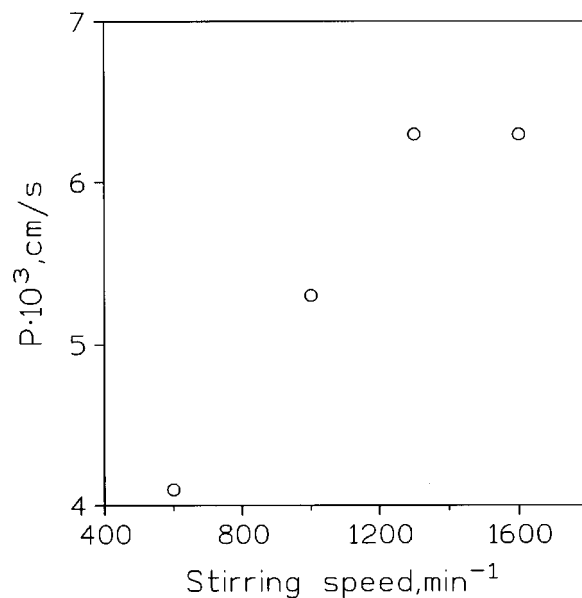
Experiments were performed to establish adequate hydrodynamic conditions. The permeability of the membrane was studied as a function of the stirring speed on the feed phase solution side as previous tests had shown that the variation of the stirring speed on the receiving solution side had no influence on gold(III) transport.

Results obtained are shown in Fig. 3. Constant permeability for stirring speeds higher than  $1300 \text{ min}^{-1}$  was obtained. Consequently, the thickness of the aqueous diffusion layer and the aqueous resistance to mass transfer were minimized. The diffusion contribution of the aqueous species to the mass-transfer process is assumed to be constant. Stirring speeds of  $1300 \text{ min}^{-1}$  and  $1600 \text{ min}^{-1}$  were maintained throughout all the experimental work for the feed and receiving phases, respectively.

### Influence of the Initial Gold and Hydrochloric Acid Concentrations

A series of experiments was performed using feed solutions with different contents of gold and hydrochloric acid, which varied from  $5 \times 10^{-5}$  to  $1 \times 10^{-3}$  M for gold and 1.0 M for HCl. The organic phase contained  $1.3 \times 10^{-1}$  M Cyanex 923 in *n*-decane.

Table 6 shows the variation of gold permeability and flux (J) for different gold concentrations. This shows that the increase of the initial gold concentration increased the metal flux but at higher gold concentrations (in the  $4 \times 10^{-4}$  M range), the flux became independent of the initial metal concentration. This may be due to membrane saturation



**Figure 3.** Influence of stirring speed on permeability of gold(III). Feed phase:  $1.3 \times 10^{-4}$  M gold in 1.0 M HCl. Membrane phase:  $1.3 \times 10^{-1}$  M Cyanex 923 in *n*-decane on a Durapore GVHP4700 support. Receiving phase: NaSCN (0.5 M) and NaCl (0.5 M).

and lower effective membrane area of the liquid membrane and also to maximization due to saturation of the membrane interface, which assists in the retention of the separating constituent on the entry side and leads to a constant flux.<sup>[24–26]</sup>

**Table 6.** Influence of initial gold concentration on gold permeability and metal flux.

[Au(III)] <sub>0</sub> (M)	P (cm/s)	J (mol/cm <sup>2</sup> s)
$5.1 \times 10^{-5}$	$6.5 \times 10^{-3}$	$3.3 \times 10^{-10}$
$1.3 \times 10^{-4}$	$6.2 \times 10^{-3}$	$8.0 \times 10^{-10}$
$2.5 \times 10^{-4}$	$6.0 \times 10^{-3}$	$1.5 \times 10^{-9}$
$3.8 \times 10^{-4}$	$5.5 \times 10^{-3}$	$2.1 \times 10^{-9}$
$5.1 \times 10^{-4}$	$4.3 \times 10^{-3}$	$2.2 \times 10^{-9}$
$1.0 \times 10^{-3}$	$2.2 \times 10^{-3}$	$2.2 \times 10^{-9}$

Membrane support: Durapore GVHP4700. Receiving phase: NaSCN (0.5 M) and NaOH (0.5 M).

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The variation in transport, as a function of the initial acid concentration, at  $1.3 \times 10^{-3}$  M Cyanex 923 in *n*-decane in the organic phase, was been studied. The experimental conditions were: aqueous feed phases containing  $1.3 \times 10^{-4}$  M gold at different HCl concentrations and receiving phases of sodium thiocyanate ( $5 \times 10^{-1}$  M) and sodium chloride ( $5 \times 10^{-1}$  M).

Results are shown in Table 7, in which an increase in gold transport occurs as the initial HCl concentration increases up to  $5 \times 10^{-1}$  M, the range at which the maximum permeation values were obtained. The reason for this behavior can be attributed to the fact that Au(III) in the feed phase is in the form of  $\text{HAuCl}_4$ ,<sup>[27]</sup> and because this metallic species is the form in which Cyanex 923 extracts gold(III) from HCl solutions.<sup>[23]</sup>

**Influence of the Organic Diluent**

To study the influence of the organic diluent on gold(III) transport, feed phases containing  $1.3 \times 10^{-4}$  M gold in 1.0 M HCl and organic solutions containing  $1.3 \times 10^{-1}$  M Cyanex 923 in each diluent were used. The results show (Table 8) that *n*-decane and cumene are the diluents with the best permeability values, compared with xylene or toluene.

**Influence of the Carrier Concentration on Permeability of Gold**

The results concerning transport of gold(III) from a feed phase containing  $1.3 \times 10^{-4}$  M Au(III) in 1.0 M HCl, the receiving phase sodium thiocyanate ( $5 \times 10^{-1}$  M), and sodium chloride ( $5 \times 10^{-1}$  M), and varying concentrations of Cyanex 923 in the range of  $1.3 \times 10^{-1}$  to  $1.3 \times 10^{-2}$  M dissolved in *n*-decane and on a Durapore GVHP4700 support, revealed no change in

**Table 7.** Influence of HCl concentration on gold permeability.

[HCl] (M)	P (cm/s)
$1.3 \times 10^{-1}$	$4.9 \times 10^{-3}$
$2.5 \times 10^{-1}$	$5.4 \times 10^{-3}$
$5.0 \times 10^{-1}$	$6.2 \times 10^{-3}$
$7.5 \times 10^{-1}$	$6.3 \times 10^{-3}$
$1.0 \times 10^{-1}$	$6.3 \times 10^{-3}$

Membrane support: Durapore GVHP4700.

**Table 8.** Influence of the diluent of the membrane phase on Au(III) transport.

Diluent	P (cm/s)
Xylene	$4.2 \times 10^{-3}$
Cumene	$5.1 \times 10^{-3}$
Toluene	$1.5 \times 10^{-3}$
<i>n</i> -Decane	$6.2 \times 10^{-3}$

Membrane support: Durapore GVHP4700.  
Receiving phase: NaSCN (0.5 M) and NaCl (0.5 M).

the permeation coefficient within these carrier range of concentrations. Accordingly, constant permeability value  $P_{\text{lim}} = 1/\Delta_{\text{aq}} = 6.3 \times 10^{-3}$  cm/s, and the thickness of the aqueous diffusion film is estimated as  $1.6 \times 10^{-3}$  cm.

### Stripping

The influence of the stripping or receiving solution on gold transport was studied using sodium thiocyanate solutions as gold strippants, due to the strong complex ability of  $\text{SCN}^-$  toward gold(III).<sup>[12,28]</sup> Experimental conditions were resumed as feed solution:  $1.3 \times 10^{-4}$  M gold in 1.0 M HCl; membrane phase:  $1.3 \times 10^{-1}$  M Cyanex 923 in *n*-decane on a Durapore GVHP4700 support; and receiving phase:  $1 \times 10^{-1}$  to  $5 \times 10^{-1}$  M NaSCN and NaCl (at constant  $I = 1$ ). Results obtained showed that under the present conditions, the concentration of sodium thiocyanate does not influence Au(III) permeability coefficient ( $6.3 \times 10^{-3}$  cm/s).

### CONCLUSION

From the experimental work performed, it can be concluded that it is feasible to use Cyanex 923 to transport gold cyanide at alkaline pH values. In this medium, the transport is not influenced by the organic phase diluent but it is influenced by the presence of lithium cations, which enhance the transport. The counter anion ( $\text{NO}_3^-$ ,  $\text{Cl}^-$  or  $\text{SO}_4^{2-}$ ) also has an influence in the transport of the precious metal. As receiving phase, either water or diluted alkaline solutions can be used to strip gold. From experimental data, the mass-transfer

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coefficient and the thickness aqueous boundary layer were estimated as  $3.6 \times 10^{-3} \text{ cm s}^{-1}$  and  $2.8 \times 10^{-3} \text{ cm}$ , respectively.

From chloride media, this reagent can also be used to transport gold at acidic feed solutions, enhancing the transport as the initial aqueous acidity is increased. The organic diluent also influences the permeation of the metal; whereas, the metal flux is also dependent on lower initial gold concentrations but becomes independent of the initial metal concentration at higher gold concentrations. Sodium thiocyanate solutions can be used effectively as receiving media. The mass transfer coefficient was calculated as  $6.3 \times 10^{-3} \text{ cm s}^{-1}$  and the thickness of the aqueous boundary layer as  $1.6 \times 10^{-3} \text{ cm}$ .

**NOMENCLATURE**

$d_{\text{aq}}$	Thickness of the aqueous feed boundary layer
$D_{\text{aq}}$	Average aqueous diffusion coefficient of the gold-containing species
$J$	Permeability flux
$P$	Metal permeability coefficient
<i>Greek Letters</i>	
$\Delta_{\text{aq}} = d_{\text{aq}}/D_{\text{aq}}$	Transport resistance due to diffusion by the aqueous-feed boundary layer

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