Application of solubility data on a hollow fiber supported liquid membrane system for the extraction of gold(I) cyanide from electronic industrial wastewater

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Abstract

The solubility ternary system was used to develop a hollow fiber-supported liquid membrane process for the extraction of gold(I) cyanide from electronics industry wastewater. Five kinds of diluents (p-xylene, cyclohexene, 1-dodecanol, n-heptane, and n-dodecane) were applied to the solubility of ternary systems (Aliquat336 + water + diluents) at 303.2 K and atmospheric pressure. 1-dodecanol, p-xylene, and cyclohexane were selected because they are completely soluble in Aliquat336 in any composition. The effects of pH on the feed solution and type of diluent were examined. The highest extraction and stripping percentages of 99.9

and 61.4, respectively, were achieved at a feed solution pH of 10.0 using p-xylene as the best diluent.

Keywords: Gold extraction; ionic liquids; solubility; hollow fiber; liquid membrane; electronic wastewater treatment

Introduction

Gold is one of the five most precious metals on earth. It is used as a very important ingredient in several industries including electronic circuits, medical, jewelry, and sensors as well as catalysts in chemical processes (Morrow et al., 2009). Gold is used in nanoparticle form for wastewater treatment due to its ability to destroy toxic materials (Fu et al., 2019) and anti-microorganisms (Adebayo-Tayo et al., 2019). Gold can exist in various forms including gold ammonia, gold thiosulfate, and gold cyanide (Cui et al., 2008). Among these, gold cyanide is the form most used in industrial processes as it has high stability (Cui et al., 2008). Potassium gold cyanide is used as a source of gold in electronic industries and released into wastewater in the form of aurocyanide ions ($Au(CN)_2^-$) (Grot, 2011; Schmidbaur and Cihonski, 2004). The highly toxic free cyanide anion (CN-) can be formed in wastewater under acidic conditions. Thus, the extraction process must be operated in a highly basic media (typically pH higher than 9.0) (Sonawane et al., 2007; Yang et al., 2018). The elimination and recovery of cyanide content from electronic industrial wastewater is of utmost importance.

Many methods have described the separation of gold(I) cyanide from solutions. Yakornov et al. used electrolytic zinc powder to separate gold by the cementation process and recorded 16.6% gold extracted from cyanide solutions (Yakornov et al., 2018). Lu Pan et al. reported 92.7% extraction of gold from a cyanide solution using 15% (v/v) furfuryl thioalcohol as the extractant and NaOH solution as the stripping phase membrane technology (Pan et al.,

2013). A good extraction percentage of 95.0% was reported by Pabby et al. using equimolar LIX79 (N,N′-bis(2-ethylhexyl) guanidine) + TOPO (trioctylphosphine oxide) by hollow fiber-supported liquid membrane contactors (Pabby et al., 2004), while Moriwaki et al. extracted gold from waste nanoparticles using an electrochemical process and obtained 35% yield (Moriwaki et al., 2017).

The hollow fiber supported liquid membrane method (Ramakul et al., 2012) was selected because it can overcome traditional solvent extraction disadvantages such as simultaneous one-step extraction and stripping. Furthermore, this method has promising features including high selectivity, low extractant (carrier) and energy consumption, as well as low capital, operating costs, large surface area to volume ratio, high mass transfer rate, and no flooding. It is also easy to scale up (Jean et al., 2018).

Trioctylmethylammonium chloride (Aliquat336) has been used as an effective extractant to extract metal ions which are in anionic form (Leepipatpiboon et al., 2013). For example, Hongmin Cui et al. reported 97.71% removal of Pb(II) ion from real waste solution using trioctylmethyl ammonium carboxymethylated chitosan ([A336][CMCTS]) (Cui et al., 2013). Hans Vigeland Lerum et al. studied the relative affinity of trioctylmethylammonium coupled with the relevant anions (perchlorate, nitrate, chloride, and sulfate) to extract cadmium from aqueous solutions (Lerum et al., 2018), while Clàudia Fontàs et al. reported the extraction of more than 50% Rh(III) from aqueous feed chloride solutions containing thiocyanate anion (SCN^-) by a hollow fiber supported liquid membrane composed of Aliquat336 (Fontàs et al., 2000). Lavinia Lupa et al. used Aliquat336 ionic liquid functionalized Zn-Al layered double hydroxide for the adsorption of phenolic compounds as other Aliquat336 applications that resulted in high adsorption efficiency (Lupa et al., 2018). Liupei Wang et al. removed more than 95% vanadium (V) from ammonium tungstate solutions by solvent extraction (Wang et al., 2018), while Li Zeng et al. reported a very high recovery

(99.0%) of Mo(VI) in the leaching solution process of spent catalysts by solvent extraction (Zeng et al., 2009). Thus, the ionic liquid Aliquat336 has proven to enable high performance in extracting metal ions in anion complex form. Therefore, Aliquat336 was used herein as the extractant to separate gold cyanide from electronic industrial wastewater by using hollow fiber supported liquid membrane technology.

The liquid membrane system has an organic liquid phase, which is a mixture of an extractant and a diluent held in a hollow fiber polymeric micropore (Schulz, 1988). Organic diluents influence the performance of liquid membrane systems. Higher mutual solubility between extractant and diluent provides increased extractability, while the lower solubility of the liquid membrane phase in aqueous solutions improves membrane stability and longevity (Leepipatpiboon et al., 2013; Parhi et al., 2012). Thus, the solubility of the extractants in diluents and solubility of diluents in the aqueous phase is very important for the liquid membrane system. Solubility is strongly related to the physical and chemical properties of each phase, especially the molecular interactions of organic solvents, extractants, and water (Bhanuprakash et al., 2016). This needs to be taken into account in the choice of diluents for the liquid membrane system.

In this work, a ternary diagram of solubility data was applied as a new method for the liquid membrane system. The optimal composition between Aliquat336, diluent, and water at 303.2 K was determined. The extraction of gold(I) ions from the effluent wastewater emanating from Mektec Manufacturing Cooperation (Thailand) Ltd was examined. The effects of the pH in wastewater, type of diluents, and membrane stability were studied.

Theory

A supported liquid membrane (SLM) consists of an organic solution of an extractant (carrier) which is held in a polymeric micropore by capillary forces (Schulz, 1988). As shown

in Figure 1, the supported liquid membrane is located between the aqueous feed solution which initially contains metal ions, and the aqueous stripping solution that initially contains no metal ions.

For the extraction mechanism, $Au(CN)_2^-$ ions initially react with Aliquat336 at the interface between the aqueous feed and liquid membrane, and then change to gold(I) complex ($R_4N^+ \bullet Au(CN)_2^-$), as shown in Eq. (1). The gold complex diffuses across the bulk liquid membrane to the interface between the liquid membrane and the stripping phase. Subsequently, the gold complex reacts with the hydroxyl group, the stripping agent in Eq. (2). Finally, the gold complex changes to $Au(CN)_2^-$ and diffuses to the bulk stripping phase, and Aliquat336 goes back to the aqueous feed solution to operate the next extraction cycle. The extraction and stripping reactions occur simultaneously. The chemical mechanism and mass transfer processes are shown in Figure 2.

$$Au(CN)_{2\,(aq)}^- + R_4N^+Cl^-_{(org)} \rightleftharpoons R_4N^+ \bullet Au(CN)_{2\,(org)}^- + Cl^-_{(aq)} \quad (1)$$

where $[R_4N^+][Cl^-]$ is Aliquat336, and $[R_3N^+ \bullet Au(CN)_2^-]$ is a gold cyanide complex. The subscripts org and aq denote the organic phases as a liquid membrane and aqueous feed phase, respectively.

$$R_4N^+ \bullet Au(CN)_{2\,(org)}^- + OH^-_{(aq)} \rightleftharpoons R_4N^+OH^-_{(org)} + Au(CN)_{2\,(aq)}^- \quad (2)$$

For the first cycle of extraction, Cl^- dissociates from the Aliquat336 molecule in the liquid membrane and diffuses to the feed phase. After that, Cl^- in the Aliquat336 molecule is replaced by OH^- from NaOH in the stripping phase. Therefore, the extractant molecule is $[R_3N^+][OH]$, while the second extraction reaction is shown as Eq. 3. The driving force of the transportation is the difference in the concentration of OH^- between the feed and stripping phases.

$$Au(CN)_{2\,(aq)}^{-} + R_4N^+OH^-_{(org)} \rightleftharpoons R_4N^+ \bullet Au(CN)_{2\,(org)}^{-} + OH^-_{(aq)} \quad (3)$$

The percentage of extraction (%E) of gold(I) ions can be defined as:

$$\%E = \frac{C_{Au,f,0} - C_{Au,f,t}}{C_{Au,f,0}} \times 100 \quad (4)$$

where $C_{Au,f,0}$ and $C_{Au,f,t}$ are the concentrations of gold(I) ions in the feed solution at the initial and final time, respectively. The percentage of stripping (%S) of gold(I) ions is defined as:

$$\%S = \frac{C_{Au,S}}{C_{Au,f,0} - C_{Au,f,t}} \times 100 \quad (5)$$

where $C_{Au,S}$ is the concentration of gold(I) ions in the stripping phase at the final time.

Experimental

Chemicals and reagents

Industrial wastewater from Mektec Manufacturing Corporation (Thailand) Ltd. was applied as a feed solution. For the liquid membrane, Aliquat336 (methyltrioctyl ammonium chloride) purchased from Sigma-Aldrich was used as the extractant and mixed with diluents including 1-dodecanol, cyclohexane, n-heptane, n-dodecane, and p-xylene. Analytical-grade potassium hydroxide was used as the stripping solution, while deionized water was used to prepare the reagent solutions. All chemical agents are listed in Table 1, with the structure of the extractant Aliquat336 shown in Figure 3.

Apparatus

Hollow fiber manufactured by Hoechst Celanese, Charlotte, NC (Liqui-Cel Extra-Flow module) was used as supporting material (Figure 1). This module consisted of Celgard microporous polypropylene fibers woven into fabric and wrapped around a central tube feeder that supplied the shell-side fluid. The woven fabric allowed more uniform fiber spacing, leading to higher mass transfer coefficients than those obtained with individual fibers (Henley et al., 2011). The properties of the hollow fiber module are specified in Table 2. The fiber was potted into a solvent-resistant polyethylene tube sheet with a shell casing of polypropylene.

Procedure

Solubility studies

Solubility data of the ternary mixtures systems were determined by the turbidimetric titration method (Yang et al., 2018). The experiment was carried out in a 15 mL glass cell under an isothermal condition of 303.2 K controlled by a water jacket. First, binary (Aliquat336 + diluents) mixtures of known compositions were prepared in the glass cell using a digital analytical balance (Sartorius TE214S with an accuracy of 0.0001 g). The composition of the binary mixture was varied at 10, 20, 30, 40, 50, 60, 70, 80, and 90% by weight, while 0 and 100% by weight mean pure diluents and pure Aliquat336, respectively). Afterward, the binary mixtures were slowly titrated with deionized water in a glass vessel at 303.2 K until mixture turbidity was observed as the equilibrium state of water solubility in the organic phase. The weight of added water was recorded and calculated to mass fraction for the ternary liquid-liquid system (extractant/diluent/water). All experiments were performed in triplicate, and the deviation of the average mass fraction value was always within ± 0.01.

Hollow fiber-supported liquid membrane

Liquid membranes (LMs) were prepared by dissolving 2.0 %v/v of Aliquat336 in a diluent, and selected as a suitable composition from the solubility data. The hollow fiber-

supported liquid membrane was operated by flowing the liquid membrane (2.0 %v/v of Aliquat336 in a diluent) through the hollow fiber module. After the hollow fiber was completely impregnated, the excess organic phase in the module was flushed out with deionized water. The feed solution from industrial wastewater was adjusted to a pH range between 9.0 and 11.0 (Sonawane et al., 2007; Yang et al., 2018) because free cyanide (CN^-) in the waste solution occurred under acidic conditions. The KOH concentration in the stripping phase was 0.5 mol/L. The feed and stripping solutions were passed through the hollow fiber in a counter-current and one-through mode using a gear pump and stirred continuously in the vessel while both solutions flowed. The flow rate of both phases was 100 mL/minute. When a steady state was reached, the samples of both outlet solutions were taken every 3 minutes. The gold(I) ion concentrations were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) (Carry 5000, Agilent Technologies, Santa Clara, CA, USA).

Results and discussion

Solubility analysis by ternary liquid-liquid system

Equilibrium solubility data for the binary liquid systems containing water in different organic agents (either Aliquat336, 1-dodecanol or cyclohexane or n-heptane or n-dodecane or p-xylene) and organic agents in the water at 303.2 K and 0.1 MPa are listed in Table 3. Solubility curves for these ternary liquid systems at 303.2 K were created and are displayed in Figure 4. The binary (Aliquat336 + n-heptane) and (Aliquat336 + n-dodecane) systems exhibited partial miscibility, while others (Aliquat336 + diluents) were completely soluble. The two liquid pairs of Aliquat336 + water and water + organic diluents were partially soluble. The systems of (a) (water + Aliquat336 + 1-dodecanol), (b) (water + Aliquat336 + p-xylene), and (c) (water + Aliquat336 + cyclohexane) systems exhibited type-2 behavior of

liquid-liquid equilibria (LLE) since the two binary pairs were partially miscible (Henley et al., 2011). The ternary system containing n-heptane and n-dodecane exhibited LLE type-3 behavior (Henley et al., 2011) since three binary pairs were partially miscible. The five kinds of organic diluents demonstrated diverse solubility results with the extractant (Aliquat336) due to differences in their molecular structure. They are aliphatic, cycloalkane, aromatic, and hydroxyl group compounds. Their structure and polarity are different and affect the solubility in Aliquat336, which can be arranged as follows: n-dodecane < heptane < cyclohexane < p-xylene < 1-dodecanol.

According to the liquid membrane's principle, the extractant and organic diluent must be most soluble for an effective liquid membrane system, while the liquid membrane should not be soluble in the water phase due to its detrimental effects on membrane stability. After considering the ternary solubility data in Figure 4 containing the five tested organic diluents, 1-dodecanol, p-xylene, and cyclohexane were found to be more suitable than n-heptane and n-dodecane. Therefore, 1-dodecanol, p-xylene, and cyclohexane were selected as diluents for the liquid membrane system to extract gold(I) cyanide from an aqueous solution.

The extraction of gold(I) using a hollow fiber-supported membrane

In this part, the one-through mode operation in Figure 1 was performed to determine the effect of pH on the feed solution and the effect of the type of diluent; the stability of the liquid membrane was also reported.

Influence of feed solution pH on the extraction of gold

The pH of the wastewater containing gold cyanide varied from 8.0 to 11.0. A pH value > 8.0 was maintained to prevent the occurrence of extremely toxic HCN from free CN^- ions in the wastewater. Figures 5 and 6 show the effects of pH in the feed solution on extraction and

stripping using three different diluents comprising 1-dodecanol, p-xylene, and cyclohexane. The extraction of gold(I) ions was enhanced with an increase in pH values and reached maximum values at pH 10 for all diluents at 99.1%, 99.9%, and 99.3% for cyclohexane, p-xylene, and 1-dodecanol, respectively.

Further increase of the pH to 11 led to a slight decrease in the gold extraction. The results demonstrated that the hollow fiber supported liquid membrane extracted gold efficiently from an aqueous solution. From the results, the percentage of stripping was relatively low, ranging from 50.0% to 65.0%, as shown in Figure 6. The stripping was enhanced when pH increased from pH 8.0 to 10.0, leading to the highest value of 61.4% with p-xylene as a diluent at a pH value of 10.0. With a further increase in pH to 11.0, the percentage of stripping value decreased to 59.0%.

Influence of organic diluent on the extraction of gold by a hollow fiber supported liquid membrane

From the ternary system in Figure 4, three diluents comprising cyclohexane, p-xylene, and 1-dodecanol were selected because they completely dissolve in Aliquat336 in any composition. From the results in Figures 5 and 6, the percentages of extraction of three diluents were practically the same at 99.9%, 99.1%, and 99.3% for p-xylene, cyclohexane, and 1-dodecanol, respectively. Similar trends were obtained for stripping at 61.4%, 59.0%, and 51.6% by using p-xylene, 1-dodecanol, and cyclohexane, respectively, as the diluent. The percentages of extraction and stripping by these three diluents were not equal because of the liquid membrane properties, especially polarity, and their solubility in water.

Table 4 shows the properties of the diluents which were used in this work (Carl, 1999). From the experimental results, both percentages of extraction and stripping of gold(I) were lower when the solubility in the water of diluent increased. It is known that the extraction and

stripping reaction occurred at the interface between the feed and liquid membrane. When the solubility in the water of diluent increases too much, the liquid membrane (diluent + extractant) is more soluble in the feed and stripping aqueous phases (see Figure 1). Therefore, the liquid membrane can easily be peeled and leaked out from the pore of the hollow fiber by the flow of aqueous feed and stripping phase. In addition, both the hydrogen bond donor and acceptor propensities of the diluents also affected the extraction process (Mohdee et al., 2020).

Stability of the liquid membrane in hollow fiber module

Figures 7 and 8 illustrate the relationship between the extraction and stripping with time in three different diluents. It can be seen that a steady-state was reached in a very short period (5 minutes), while the percentage of extraction was constant after 8 minutes. The stability of the liquid membrane can be determined if the extraction and stripping values remain constant. The results showed that the liquid membrane stability was more than 120 minutes.

5. Conclusions

Cyclohexane, dodecanol, and p-xylene were selected as the diluents in a hollow fiber-supported liquid membrane system because they are completely soluble in Aliquat336 at any composition. The highest extractability was obtained at a pH of 10.0. The highest percentages of extraction and stripping were achieved by using p-xylene as a diluent. More than 99.0% of gold(I) could be recovered from industrial wastewater.

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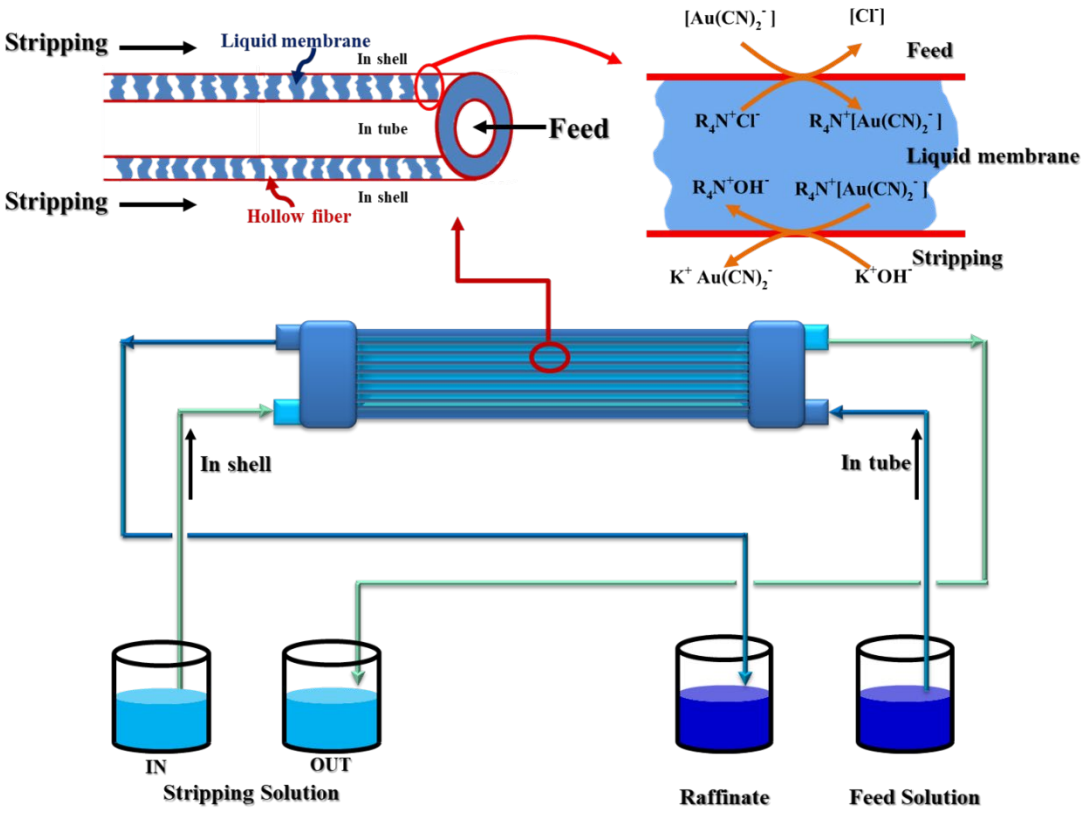

Fig. 1 Hollow fiber supported liquid membrane

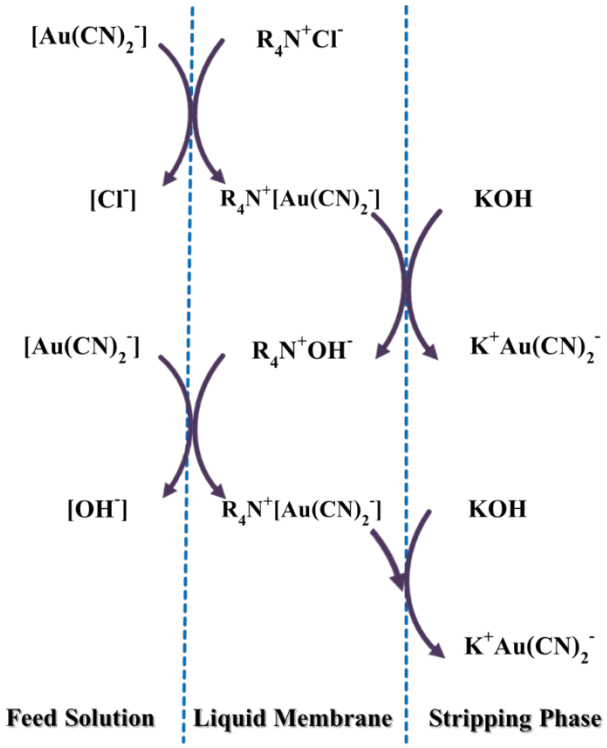

Fig. 2 Extraction and stripping reaction mechanism

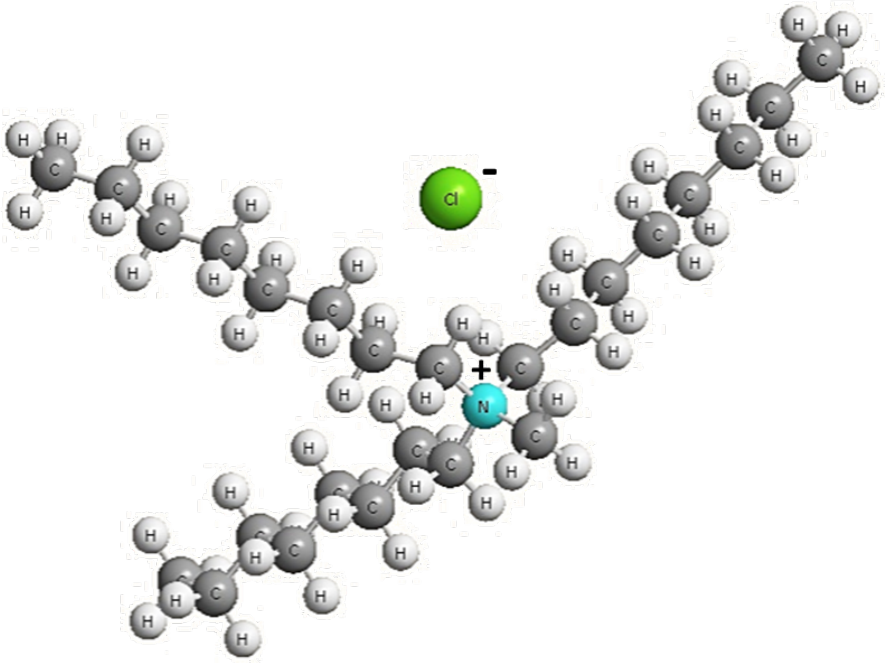

Fig. 3 3D molecular structure of Aliquat336

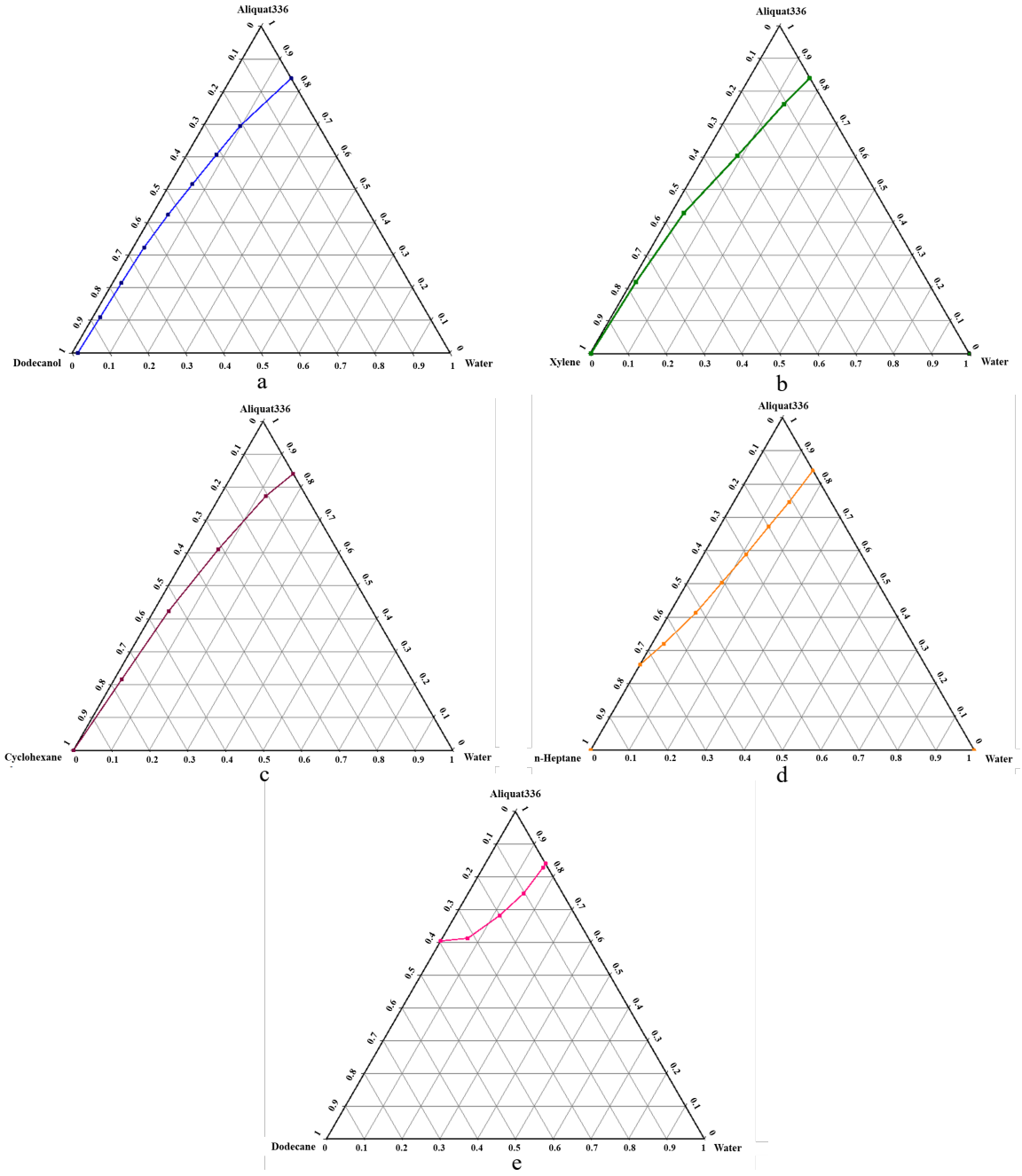

Fig. 4 Solubility diagram for the system containing Aliquat336, water and organic diluents (1-dodecanol(a), xylene(b), cyclohexane(c), n-heptane(d), n-dodecane(e)) at 303.2 K and 0.1 MPa in different mass fraction

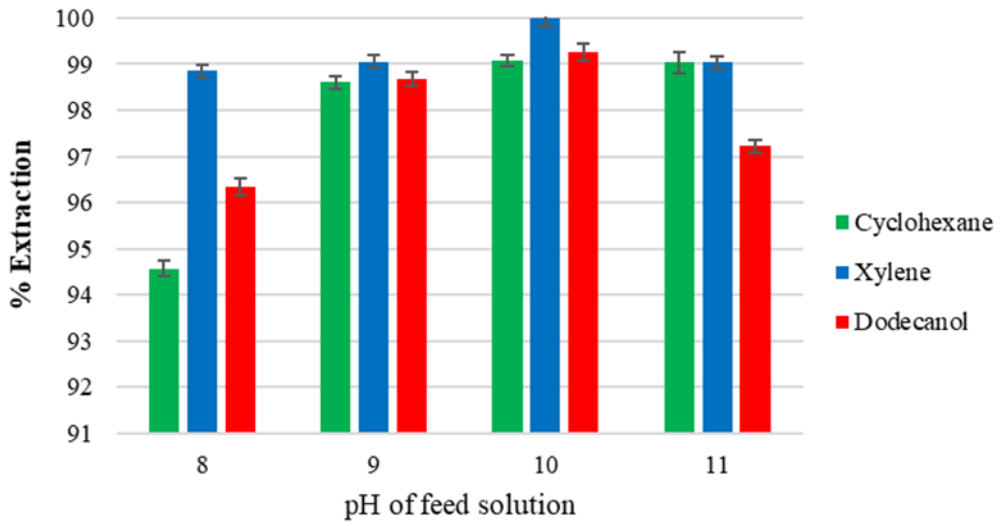

Fig. 5 Relation between the extraction of gold(I) cyanide and pH of the feed solution in different diluents at 303.2 K

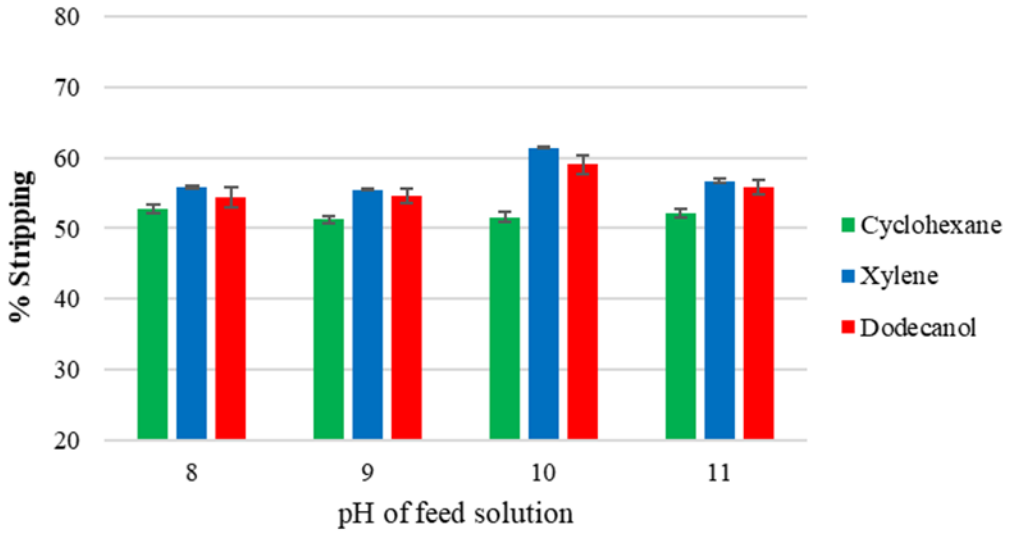

Fig. 6 Relation between the stripping of gold(I) cyanide and pH of the feed solution in different diluents at 303.2 K

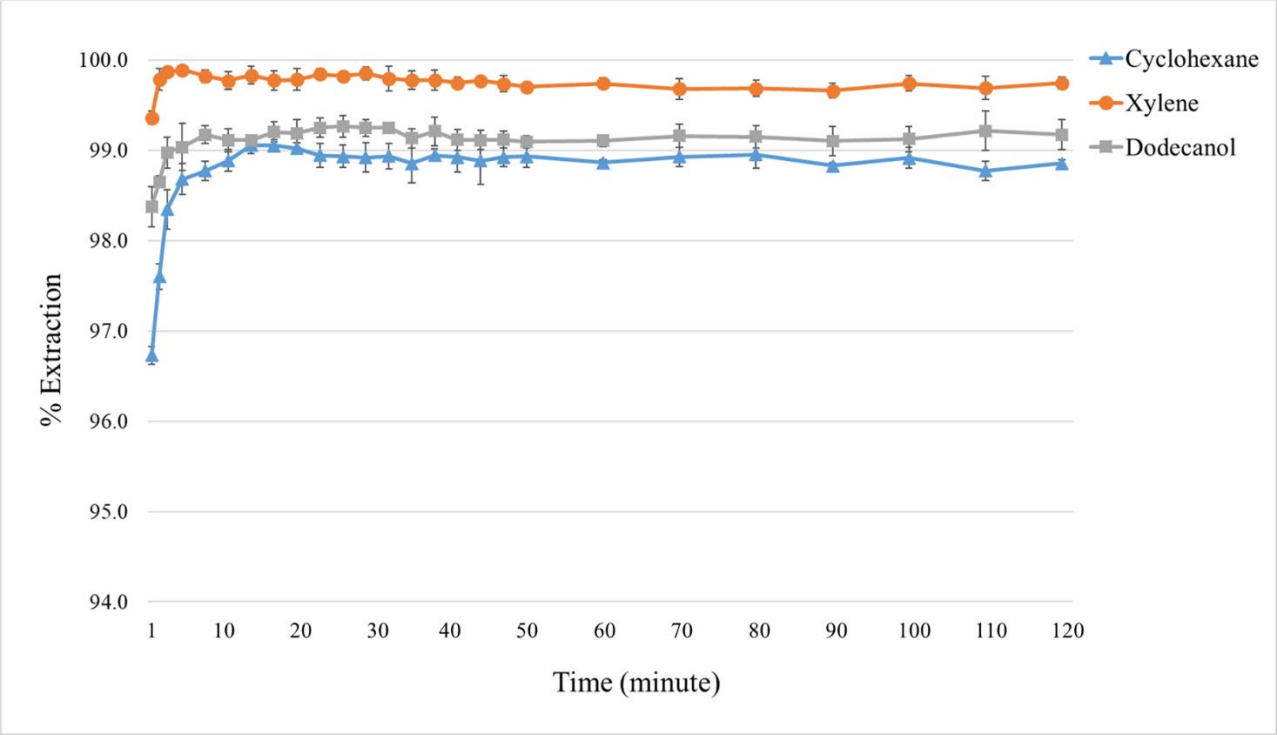

Fig. 7 Relation between the extraction of the gold(I) cyanide and time in diluent (at pH feed solution of 10.0) at 303.2 K

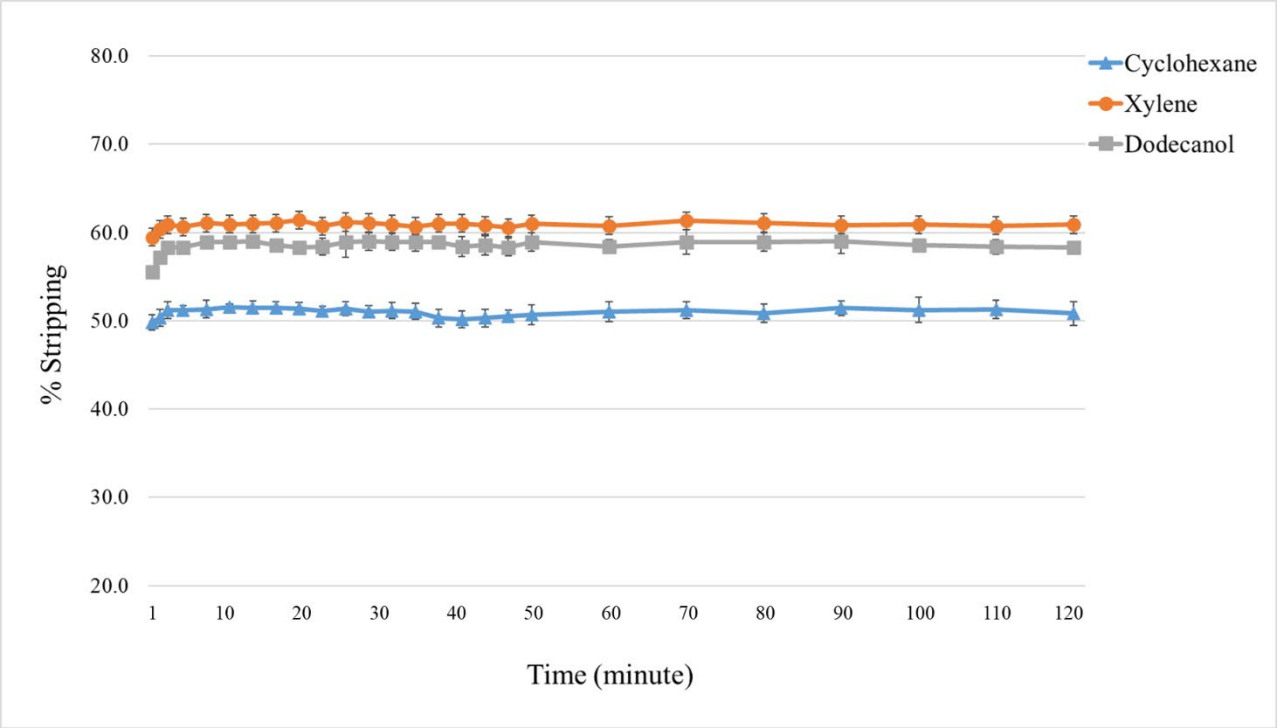

Fig. 8 Relation between the stripping of the gold(I) cyanide and time in diluent (at pH feed solution of 10.0) at 303.2 K

Table 1. Chemical reagents

Chemicals	**Molecular Formula**	**Molecular weight (g/mol)**	**Purity %(w/w)**
Trioctylmethylammonium chloride (Aliquat336)	$C_{25}H_{54}Cl$	404.2	≥90.6
1-Dodecanol	$C_{12}H_{26}O$	186.3	≥98.0
Cyclohexane	C_6H_{12}	84.2	≥99.7
n-Heptane	C_5H_{12}	100.2	≥99.2
n-Dodecane	$C_{10}H_{22}$	170.3	≥98.5
p-Xylene	C_8H_{10}	106.2	≥98.5
Potassium hydroxide	KOH	56.1	≥98.0

Table 2. Properties of Hollow Fiber Module (Ramakul et al., 2012)

Properties	Description
Material	Polypropylene
Inside diameter of hollow fiber	240 μm
Outside diameter of hollow fiber	300 μm
Pore size	0.05 μm
Porosity	30%
Contact area	1.39 m^2
Area per unit volume	29.3 cm^2/cm^3
Module diameter	6.3 cm
Module length	20.3 cm

Table 3. Mutual solubility of binary systems (solubility of (1) in (2)) at 303.3 K in mass fraction

		(2)						
		Water	**Aliquat 336**	**1-Dodecanol**	**Cyclohexane**	**p-Xylene**	**n-Heptane**	**n-Dodecane**
(1)	**Water**		0.1591**	0.0148**	0.0008 (Danon et al., 2018)	0.00269 (Polak and Lu, 1973)	8.2E-5 (Polak and Lu, 1973)	None
	Aliquat336	0.0012**		/	/	/	0.2669**	0.6045**
	1-Dodecanol	4.29E-06 (Amidon et al., 1974)	/		/	/	/	/
	Cyclohexane	5.58E-05 (Danon et al., 2018)	/	/		/	/	/
	p-Xylene	1.70E-04 (Polak and Lu, 1973)	/	/	/		/	/
	n-Heptane	3.37E-06 (Amidon et al., 1974)	0.7331**	/	/	/		/
	n-Dodecane	3.80E-09 (Sutton and Calder, 1974)	0.3955**	/	/	/	/	

(/) Completely soluble
** Experiment results from this work

Table 4. Properties of the diluents used in this work (Carl, 1999)

Diluents	Density (g/mL at 25°C)	Viscosity (cP at25 °C)	Polarity index (-)	Solubility in water (mg/L at 25 °C)
p-Xylene	0.876	0.617	2.5	2.010
1-dodecanol	0.833	16.44	N/A	4.000
cyclohexane	0.779	0.903	0.2	5.610
dodecane	0.75	1.19 cP	0.0	0.003
heptane	0.684	0.391	0.1	2.200