Application of Experimental Design to Emulsion Liquid Membrane Pertraction of Gold (III) Ions from Aqueous Solutions

A. Kargari, T. Kaghazchi*, M. Sohrabi, M. Soleimani Department of Chemical Engineering- Amirkabir University of Technology (Tehran Polytechnic) – Tehran, Iran.

Abstract

The application of the Taguchi analysis to the experimental results of the extraction of gold (III) ions by a three-phase W/O/W emulsion liquid membrane system has been studied. Experiments were performed using alkaline aqueous sodium sulfite as the internal phase and LK-80 as a novel emulsifier for the first time in emulsion liquid membrane technology. The effects of various operational and compositional parameters were studied. Results showed that nearly all of the Au (III) ions in the aqueous phase were extracted in a few minutes. The analysis determined the effects and contribution of each of the parameters on the extraction efficiency, interaction between the parameters and also the optimum parameters to achieve the best extraction efficiency. The analysis showed that the pH of the external phase with a contribution of 65% is the highest, and the concentration of carrier with a contribution of 2% is the least effective in the final system response.

Keywords: emulsion liquid membrane- gold – extraction- carrier - Taguchi analysis

Introduction

The Taguchi method [1,2] is a systematic application of design and analysis of experiments for the purpose of designing and improving product quality. In recent years, the Taguchi method has become a powerful tool for improving productivity during research and development so that high quality products can be produced quickly and at low cost.

Optimization of process parameters is the key step in the Taguchi method in achieving a high quality without increasing the cost. This is because the optimization of process parameters can improve performance characteristics and the optimal process parameters obtained from the Taguchi method are insensitive to the variation of environmental conditions and other noise factors. Basically, classical process parameter design is complex and not easy to use, especially as a large number of experiments have to be carried out when the number of the process parameters increases. To solve this task, the Taguchi method uses a special design of orthogonal arrays to study the entire process parameter space with a small number of experiments only. A loss function is then defined to calculate the deviation between the experimental value and the desired value. Taguchi recommends the use of the loss function to measure the performance characteristic deviation from the desired value. The value of the loss function is further transformed into a

^{* -} E-mail: kaghazch@aut.ac.ir

signal-to-noise (S/N) ratio.

Usually, there are three categories of performance characteristic in the analysis of the S/N ratio, i.e. the lower-the better, the higher-the-better, and the nominal-the-better. The S/N ratio for each level of process parameters is computed based on the S/N analysis. Regardless of the category of the performance characteristic, a larger S/N ratio corresponds to a better performance characteristic. Therefore, the optimal level of the process parameters is the level with the highest S/N ratio. This is true for the optimization of a single performance characteristic.

Extraction by emulsion liquid membranes

The idea to perform extraction in a threephase system with emulsion liquid membranes (ELM's) is relatively new. Emulsion liquid membranes technology was first proposed by N.N. Li for the separation of hydrocarbons [3-5]. From then on, several applications have been developed concerning the removal and recovery of several solutes from either aqueous or organic solutions including separation of hydrocarbons [5], recovery and removal of organic acids and amines [6,7], waste water treatment [8-11], recovery and purification of metal ions [10-12] and biomedical applications such as artificial kidney [7].

In this technology, solutes are not only removed, but also concentrated: extraction and stripping are carried out in a single step. A schematic diagram of this technology is presented in Figure 1.

Emulsion liquid membranes are typically made by, first dispersing the internal phase in an immiscible liquid and then dispersing this emulsion in a third phase (called the external phase). Normally, the internal and the external phases are miscible, but both are immiscible with the membrane phase separating them [13,14].

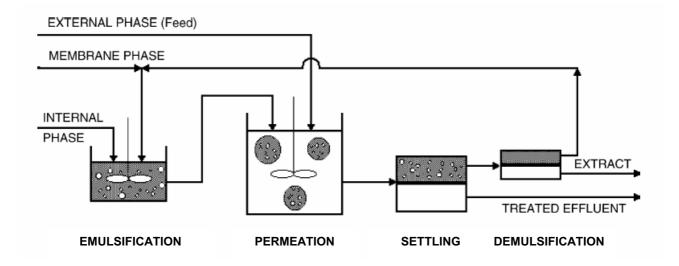


Figure 1. Schematic of a continuous emulsion liquid membrane process

The effluent to be treated is contacted with an emulsion dispersed in globules (with diameters in the 0.1-2mm range). Each globule consists of droplets of an aqueous internal receiving phase (with diameters in the 1-10 um range) encapsulated in a membrane phase containing a surfactant (Figure 2). The emulsion was previously prepared by stirring at high mixing speed (emulsification stage). Depending on the nature of external and internal phases, the emulsion can be W/O or O/W type, so the three-phase extraction system is either O/W/O or W/O/W type. During the contact between wastewater and emulsion globules (permeation), solute transport occurs through the membrane and into the internal receiving phase, where it is concentrated. After the subsequent separation of the effluent from the emulsion by gravity (settling), splitting of the emulsion (demulsification) is carried out. Demulsification is usually performed by applying an electrical field, in order to separate the phases, which make up the emulsion-the internal aqueous phase and the organic membrane phase. At the end of the process, the membrane phase can be reused and the receiving phaseenriched in the recovered solute-can be recycled or recovered for solute. According to its composition, the effluent may be submitted to additional treatment processes or simply discharged.

Five industrial plants of emulsion liquid membranes have already been reported in the literature: four applied to zinc removal from wastewater of the viscose fiber industry and one applied to the treatment of phenolic waste water of a plastic factory in the People's Republic of China [13].

Besides the referred industrial application, several batch studies with phenols are known. The majority has been performed with synthetic aqueous solutions of phenol [15,9], but there are also studies using aqueous solutions of other phenols, such as cresols, chlorophenols and nitrophenols [16-20]. Lack of work with industrial effluents including modeling is, however, still noticeable.

Solvent extraction has been widely used in the recovery of metals from aqueous solutions [21]. In these processes, metal ion containing solution makes contact with a large amount of selective solvent. After extraction, this process is followed by stripping. Solvent extraction is very difficult for the separation of trace amounts of metal ions because of low driving force, and also a large amount of solvent is required. These make the extraction and stripping of the desired species very expensive. Extraction of metal ions by ELM systems has been considered as a powerful alternative for conventional solvent extraction systems. Due to highly expensive selective solvents for gold, gold extraction using liquid-liquid extraction is not economical.

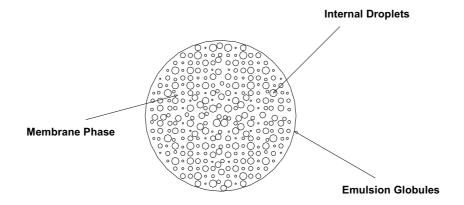


Figure 2. A schematic diagram of emulsion liquid membrane (emulsion globule)

A brief comparison of liquid-liquid extraction and liquid pertraction has been done [22]. It follows from the above that liquid pertraction is a combination in time and space of two well-known separation process-ses-solvent extraction and solvent stripping. This combination offers two substantial advantages over the classical liquid-liquid extraction:

- Pertraction provides maximum driving force so that the use of multistage and counter-current processes is not required.
- Since the organic liquid is a short- term mediator only, its extraction capacity is of no essential significance. As a result, a great variety of insoluble, inert and harmless organic liquids can be used as an intermediate liquid, containing only a small amount of carrier. As a carrier, new, highly selective and also solid extractants can be synthesized.

In the previous works, separation of hydrocarbons, organic acids, amines, some metal ions, wastewater treatment, etc. has been studied, but little attention was paid to the separation of gold ions. Several techniques for gold extraction from aqueous solutions have been invented such as solvent extraction, carbon adsorption, ion exchange, cementation, etc. [21]. In the case of conventional solvent extraction technique for the recovery of gold from hydrochloric acid solutions, several studies have been performed using various types of extractants such as basic extractants like amines, solvating extractants such as neutral organophosphorus compounds and other extractants containing S as the donor atom. Few studies have been done on the separation of gold ions by liquid membranes [21, 23-25]. Most of them were done by using supported liquid membranes [23]. The only study of gold extraction by emulsion liquid membrane was carried by Yan et.al. [26].

In the present study, a novel emulsifier was used for the preparation of the desired emulsions, which is very stable to highly alkaline-reducing and acidic aqueous solutions. The emulsifier has a natural base and so is biodegradable. We named it LK-80. Chemically, it is a wax, being a complex mixture of esters and polyesters of 33 high-molecular-weight alcohols and 36 fatty acids. In this work, methyl iso-butyl ketone (MIBK) was used as the carrier and liquid paraffin as the membrane phase.

Experimental

Apparatus

All experiments were performed at a constant temperature of 25±0.2°C using a water bath equipped with a P- type controller and an agitator. Extraction experiments were carried out in a cylindrical glass container with 105mm diameter and 145mm height equipped with four round glass baffles with a 8mm diameter. The stirrer for the extraction experiments was made of glass with a 43mm diameter and had four pitched blades with a 30° angle. The stirring driver was a laboratory mixer model RE-166 from Ika-Werk Germany with a variable speed of 50-6000 rpm. The stirrer for the emulsion preparation was made from stainless steel and coated with LDPE to minimize the contamination of the emulsion. It had six pitch blades with 45° angle and 63mm diameter.

Sampling were done at predetermined time intervals by disposable sanitary syringes and centrifuged by a laboratory centrifuge in order to separate the emulsion phase from the external phase. To determine the concentration of gold ions in the external phase, an atomic absorption spectrophotometer (Unicam model 939) was used. pH measurements were carried out using a Teb Azma pH meter.

Reagents

Sodium hydroxide (NaOH), sodium sulfite (Na₂SO₃), iso-butyl methyl ketone ((CH₃) ₂CHCH₂COCH₃), nitric acid (HNO₃) and auric chloride (Au HCl₄.4H₂O) were of reagent grade and purchased from Merck.

market. The HLB value for LK-80 was 4.5. Liquid paraffin with a density of 0.82-0.84 gr/cm³ at 20°C and a viscosity of 25-26 cP at 40°C was supplied from Rose Polymer Co. Stock gold chloride solutions were prepared by dissolution of pure metallic gold with 99.99 assays in a mixture of hydrochloric acid and nitric acid with a defined volumetric ratio (aqua regia). The solution that completely dried by evaporation was then dissolved in a mixture of hydrochloric acid and distilled water with a defined proportion. Distilled water was used throughout.

Emulsifier LK-80 was purchased from local

Procedure

All experiments were performed at 25±0.2°C. Emulsions were prepared by mixing the liquid paraffin and the emulsifier under moderate heating. Then, the internal phase was added to the mixture and mixed for 10 minutes at 6000rpm. Finally, the carrier was added to the emulsion and mixed for an additional 2 minutes. The final volume of the emulsion was about 137 ml.

The external phases were prepared by diluting the stock solution with distilled water to 500ml followed by pH adjustment. The external phases were placed into a water bath for 20 minutes and then the emulsion was added to the external phase with stirring. The external phase solution was periodically sampled at certain time intervals. The concentration of gold in the solution was determined after separation of the emulsion phase, by the atomic absorption spectrophotometer at 242.8 nm.

Results and Discussion

Kinetic of pertraction

It is well established that an anion carrier can act as a selective carrier for chemical species in transportation processes [27]. In chloride solutions, gold exists as complex anion $AuCl_4^-$. MIBK molecule presents oxygenunpaired electrons, so they tend to unite with

protons (H⁺) in acidic solution and then react with $AuCl_4^-$. The carrier MIBK extracts gold on the external surface of emulsion droplets according to the following reactions:

$$AuHCl_4.4H_2O + H_2O \rightarrow AuCl_4^- + 5H_2O + H^+$$
(1)

$$n(C_4H_9 - (CH_3)C = O:)_{org} + H_{aq}^{+} \rightarrow$$

$$\left[(C_4H_9 - (CH_3)C = O:)_n H \right]_{aq}^{+}$$
(2)

$$[(C_{4}H_{9} - (CH_{3})C = O:)_{n}H]_{org}^{+} + AuCl_{4 aq}^{-} \rightleftharpoons$$

$$([(C_{4}H_{9} - (CH_{3})C = O:)_{n}H]^{+} AuCl_{4}^{-})_{org}$$
(3)

The gold-MIBK complex transfers across the membrane phase and reacts with the internal phase at the outer surface of internal phase droplets according to the following reaction:

$$\begin{split} 2Na_{2}SO_{3} + & \left(\left[(C_{4}H_{9} - (CH_{3})C = O:)_{n}H \right]^{+} AuCl_{4}^{-} \right)_{org} \\ + NaOH \rightarrow NaAuSO_{3} + Na_{2}SO_{4} + n(C_{4}H_{9} - (CH_{3})C + O:)_{org} \\ + 2HCl + 2NaCl \end{split} \tag{4}$$

According to the above reaction, Au³⁺ reduces to Au⁺ in the alkaline sulfite solution. At the extraction condition, Au⁺ ions are not able to form a complex formation with MIBK and move back to the external solution [26].

As the stoichiometric relation for the MIBK-gold complex formation was considered as equation of (3), then:

$$K_{ex} = \frac{[(MIBK)_{n}H^{+}AuCl_{4}^{-}]_{org}}{[MIBK]_{org}^{n}[H^{+}]_{aq}[AuCl_{4}^{-}]_{aq}}$$
(5)

The experimental runs were performed at 25° C, using various MIBK concentrations within the organic phase. Samples were taken from the aqueous phase until equilibrium conditions were established. The concentration of $AuCl_4^-$ in the aqueous phase was measured. The concentrations of the latter within the organic phase were determined from the differences between the initial concentrations of $AuCl_4^-$ and those measured in the aqueous phase.

From Figure 3, constructed by plotting the $\log \frac{[(MIBK)_n H^+ AuCl_4^-]_{org}}{[H^+]_{ag} [AuCl_4^-]_{ag}}$ vs. $\log [MIBK]_{org}$,

it may be observed that a linear curve has been formed, having a slope of 4.0081 and intercept of -1.0034, then, n=4 and $K_{ex} = 0.09922 \ (dm^3/mol)^5$.

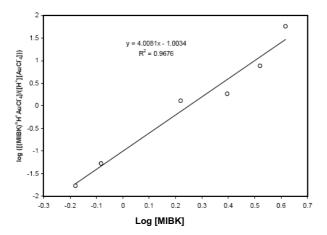


Figure 3. Equilibrium extraction of Au³⁺ by MIBK

Consequently, the stoichiometric relation for the reaction may be written as follows:

$$4(MIBK)_{org} + H_{aq}^{+} + AuCl_{4aq}^{-} \rightleftharpoons [(MIBK)_{4}H^{+}$$

$$AuCl_{4}^{-}]_{org}$$
(6)

Effect of external phase concentration

An important parameter affecting the extraction/stripping process appears to be the initial Au³⁺ concentration. The Au³⁺ concentration in the source aqueous solution was varied from 21.4 to 109.6 ppm. In Figure 4, kinetic profiles of extraction (and also stripping) degrees at different initial Au³⁺ concentrations, at a speed of agitation of 510 rpm, are demonstrated.

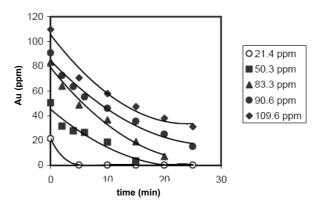


Figure 4. Effect of the external phase on the extraction efficiency $C_i = 0.4M\ Na_2SO_3,\, 0.4M\ NaOH;\, C_c = 1.1\%;\, R_{ew} = 0.274;\, pH = 0;510\ rpm.$

It is reasonable that by increasing the Au³⁺ concentration in the external phase, the time for extraction also increases. The fixed amount of carrier in the experiments causes nearly the same extraction rate. In other words, the time for extraction increases by increasing the external phase concentration. This is true for all extraction processes.

Effect of the carrier concentration

The effect of MIBK as the carrier in the membrane phase in the range of 0.365 to 2092 vol.% is shown in Figure 5.

It is well known that the rate of facilitated transport of the analytes is directly affected by the amount of carrier immobilized in the corresponding liquid membrane.

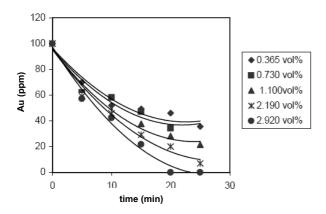


Figure 5. Effect of carrier concentration on the extraction efficiency

 $C_e = 100 \ ppm$; $C_i = 0.4M \ Na_2SO_3$, 0.4M NaOH; $R_{ew} = 0.274$; pH = 0; 510 rpm.

Increasing the amount of carrier has two effects. The viscosity of the membrane phase, which limits the extraction rate, decreases by

increasing the carrier concentration, and hence the carrier acts as a thinner for the membrane phase. At the same time, increasing the carrier concentration over a certain limit decreases the stability of the emulsion [28-32]. On the other hand, an increase in the concentration of the carrier in the membrane phase increases the ability of the membrane phase for extraction. The best value of the carrier concentration was found to be about 2.92vol%, based on the emulsion phase in our experiments.

Effect of reagent concentration in the internal phase

The effect of the internal phase concentration on the extraction efficiency has been shown in Figure 6. Increase in the internal phase reagent concentration increases the ability of the internal phase for stripping the gold-MIBK complex at the external surface of the internal droplets and reduces the concentration polarization at the surface; so the driving force for the extraction increases [13]. The results showed that the best concentration for the internal phase regent was 0.5M for sodium sulfite. These experiments were performed at a constant NaOH concentration of 0.4M and different concentrations of sodium sulfite.

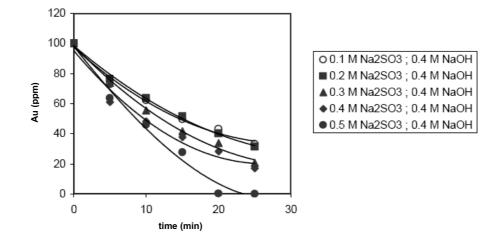


Figure 6. Effect of internal phase reagent concentration on the extraction efficiency $C_e = 100 \ ppm$; $C_c = 1.1\%$; $R_{ew} = 0.274$; pH = 0; $510 \ rpm$.

Effect of the external phase pH

The effect of the pH at the external phase on the extraction efficiency has been shown in Figure 7. Any decrease in pH increases the extraction efficiency.

This effect can be explained by Eqns. (2) and (3): the rate of the complex formation depends directly on the H⁺ concentration in the external phase, so by reducing the pH value for the extraction phase, the extraction

efficiency increases.

Effect of agitation speed

When the permeation stirring speed is increased, the shear forces, which act on the emulsion globules, increase, and then the average emulsion globules radius decreases and the interfacial area available for the mass transfer increases. Thus, the mass transfer rate increases as illustrated in Figure 8.

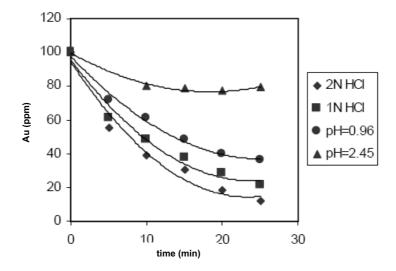


Figure 7. Effect of external phase pH on the extraction efficiency $C_o = 100 \ ppm$; $C_i = 0.4M \ Na_2SO_3$, 0.4M NaOH; $C_c = 1.1\%$; $R_{ew} = 0.274$; 510 rpm.

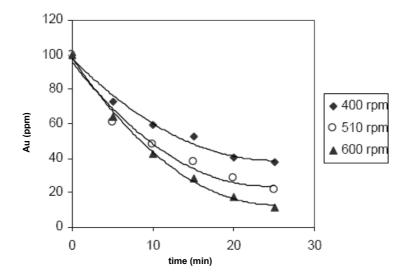


Figure 8. Effect of speed of agitation the extraction efficiency $C_e = 100 \ ppm$; $C_i = 0.4M \ Na_2SO_3$, 0.4M NaOH; $C_c = 1.1\%$; $R_{ew} = 0.274$; pH = 0.

As other authors have pointed out, the external surface area of globules rather than the internal phase droplet surface area control the extraction process with ELMs [4,13, 14,33], so by increasing the speed of agitation the area for mass transfer increases and the extraction efficiency increases. However, there is an upper limit for this effect, caused by the non-ideality phenomena in this system (namely membrane phase leakage with exit of internal phase into the external phase), thus reducing the efficiency of the whole process. A lower limit for the permeation stirring speed might also be drawn: it should be high enough to disperse the emulsion globules evenly in the external phase. This situation depends on the permeation reactor geometry [29].

Analysis of the Results

In the present study, there are eleven degrees of freedom owning to the four sets of threelevel and one level of three-level extraction process parameters. The degree of freedom for the orthogonal array should be greater than or at least equal to those for the process parameters. The extraction parameters and their levels are shown in Table 1.

In this study, a M_{16} orthogonal array with four columns and sixteen rows was used. This array has fifteen degrees of freedom and can handle four-level process parameters, with 4 degrees of freedom for the error. Sixteen experiments are required to study the entire extraction parameter space when the M_{16} orthogonal array is used.

Sixteen experiments with the arrangement of M_{16} were performed. The extraction rate was measured after 20 minutes. The extraction rate is the higher-the-better performance characteristic. The results are shown in Table 2. The effect of each extraction process parameter on the main effect at different levels can be separated out because the experimental design is orthogonal. Figures 9-12 show the response value for each level.

Table 1. Extraction parameters and their 1

Symbol	Process parameter	Unit	Level 1	Level 2	Level 3	Level 4
A	C_c	Vol%	0.365	1.100	2.190	2.920
В	C_{i}	mol/lit	0.1	0.3	0.4	0.5
C	pН	-	2N HCl	1N HCl	0.96	2.45
D	N	rpm	400	510	600	-

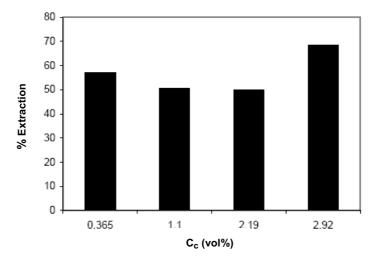


Figure 9. Main effects of the concentration of carrier on the extraction efficiency

Table 2. Experimental layout and the results for percent of extraction after 20 minutes

Experiment _ number	Process parameter level				Entroption 0/	C/N(ID)
	A	B C D		D	Extraction %	S/N ratio (dB)
1	1	1	1	1	45.60	33.18
2	1	2	2	2	93.275	39.39
3	1	3	3	3	87.22	38.81
4	1	4	4	1	2.00	6.02
5	2	1	2	3	59.50	35.49
6	2	2	1	1	52.80	34.45
7	2	3	4	1	2.00	6.02
8	2	4	3	2	87.22	38.81
9	3	1	3	1	28.10	28.97
10	3	2	4	3	6.70	16.52
11	3	3	1	2	93.975	39.46
12	3	4	2	1	70.60	36.97
13	4	1	4	2	31.30	29.91
14	4	2	3	1	87.22	38.81
15	4	3	2	1	93.21	39.39
16	4	4	1	3	61.50	35.77

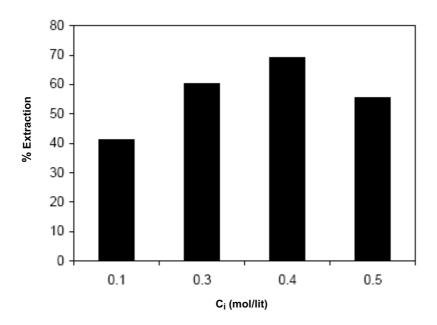


Figure 10. Main effects of the concentration of internal phase on the extraction efficiency

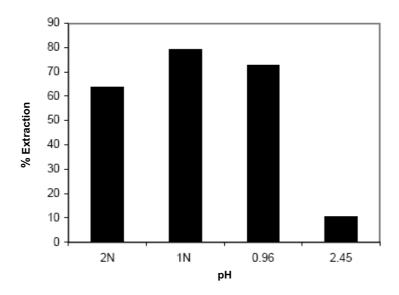


Figure 11. Main effects of the pH of external phase on the extraction efficiency

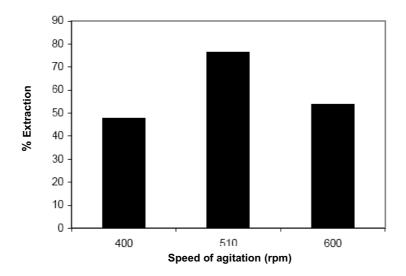


Figure 12. Main effects of the speed of agitation on the extraction efficiency

The above results show that there are some interactions between the parameters. To find the interaction between the parameters, the response of each parameter against the others must be constructed. The results of analysis are listed in Table 3.

The results of interaction show that the main

interactions are between C_c - C_i and C_c -pH. This is due to the dependence of the extraction ability of MIBK to the pH of the aqueous phase. This result is in agreement with our experimental data and Eqns. (2) and (3).

Zwaze Contraction of French Contraction of the French Contraction of t							
Process parameter	A	В	C	D			
A	-	29.55	22.27	5.53			
В	29.55	-	14.44	16.5			
C	22.27	14.44	-	18.15			
D	5 53	16.5	18 15	_			

Table 3. Results of percent of interaction between the process parameters

Basically, the larger the response, the better the performance characteristic. However, the relative importance among the extraction process parameters for the performance characteristic still needs to be known so that the optimal combinations of the extraction process parameter levels can be determined more accurately. By considering the above results, it was found that the best combination of the extraction process parameters is $A_4B_3C_2D_2$. In other words, by selection of this combination of the extraction process parameters, the best response with minimum required conditions (cost) would be attainable [34].

Analysis of variance

The purpose of the ANOVA is to investigate which extraction process parameters signify-cantly affect the performance characteristic. This is accomplished by separating the total variability of each level, which is measured by the sum of the squared deviations from the total mean of the responses, into contribution by each extraction process parameter and the error. The percentage contribution by each of the process parameters in the total sum of the squared deviations can be used to evaluate

the importance of the process parameter change on the performance characteristic. In addition, the F-test named after Fisher [35] can also be used to determine which extraction parameters have a significant effect on the performance characteristic. Usually, changing the extraction process parameter has significant effect on the performance characteristic when the F value is large.

The results of ANOVA that are shown in Table 4, indicate that the pH value of the external phase, speed of agitation and concentration of sodium sulfite in the internal phase are the significant extraction process parameters for affecting the performance characteristic. Furthermore, the pH value of the external phase is the most significant extraction process parameter due to its highest percentage contribution (65.374%) among the process parameters. Based on the above discussion, the optimal extraction process parameters are the concentration of carrier level 4, concentration of the reagent in the internal phase level 3, pH value of external phase level 2 and speed of agitation level 2 ($A_4B_3C_2D_2$).

Table 4. Results of the analysis of variance

Process parameter	DOF	Variance	F-ratio	Contribution%
Concentration of carrier (A)	3	295.203	1.75	2.213
Concentration of internal phase (B)	3	544.990	3.232	6.579
pH of ext. phase (C)	3	3,908.600	23.183	65.374
Speed of agitation (D)	2	1,121.028	6.649	11.098
Other/Error	4	168.591	-	14.736
Total	15	=	-	100.000%

By use of the above analysis, both the effects of each parameter on the extraction efficiency and the counter effects of parameters on each other can be observed. From the Fisher tables [35] with 90% and 95% confidence, $F_{0.1,3,4} = 4.19$, $F_{0.1,2,4} = 4.32$ and

 $F_{0.05,3,4}=6.59$, $F_{0.05,2,4}=6.94$. According to these values, the F value for pH and the speed of agitation are greater than the corresponding values of Fisher tables, so the tests are reliable with 90% and 95% confidence, respectively, and these parameters have great effects on the performance

characteristic of the extraction process. The contribution of the error is 14.7%, which is in a reasonable range of errors.

The experiment with the optimal conditions, which was predicted by the Taguchi analysis, was performed. The results are presented in Figure 13. The results showed that the remaining gold ions concentration in the aqueous solution, after 20 minutes, was behind the detection limit of the spectrophotometer (less than 0.1ppm), so the extraction efficiency was more than 99.9%.

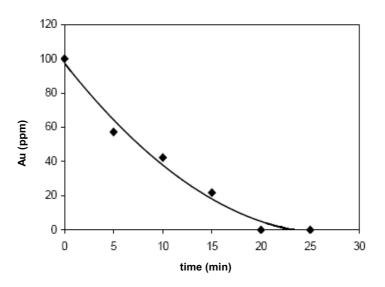


Figure 13. Result of extraction at the optimum conditions $C_e = 100 \ ppm$; $C_i = 0.4M \ Na_2SO_3$, 0.4M NaOH; $C_c = 2.92\%$; $R_{ew} = 0.3$; pH = 0; 510 rpm.

Conclusion

the application of the Taguchi method as a powerful technique for the analysis of the experimental results of gold ions extraction by ELM has been shown. The analysis of the data shows that there are some interactions between the extraction parameters and the best experimental condition, which was predicted using the Taguchi analysis is $(A_4B_3C_2D_2)$. The contributions of pH and

speed of agitation on the extraction efficiency are greater in comparison with the other parameters, whereas the contribution of carrier concentration is negligibly small. In this study a new efficient emulsifier (LK-80) has been introduced for the extraction of gold (III) ions from acidic aqueous media using MIBK as the carrier and alkaline sodium sulfite solution as the internal reagent. The extraction rate increases with the increase of

the carrier concentration, internal phase concentration, agitation speed and decrease of the external phase pH. This emulsion is capable of extracting, almost completely, the total gold ions present in the external phase within a few minutes [36].

Nomenclature

- C_c Concentration of carrier in the emulsion phase, vol%.
- C_e Concentration of gold (III) ions in the external phase, ppm.
- *C_i* Concentration of sodium sulfite in the internal phase, mol/lit.
- R_{ew} The ratio of emulsion phase to the external phase, vol/vol.
- N Speed of agitation, rpm.

References

- 1. Taguchi, G., Introduction to quality engineering, Asian Productivity Organization, Tokyo, (1990).
- 2. Peace, G.S., Taguchi methods: A hand-on approach, Addison Wesley Reading, MA, (1993).
- 3. Li, N.N., US-Patent 3410794, (1968).
- 4. Elzinga, E.R. and Li, N.N., US-Patent 3389078, (1968).
- 5. Li, N.N., Permeation through liquid surfactant membranes, *AIChE J.*, **17**, 459 (1971).
- 6. Eyal, A.E. and Bressler, E., Industrial separation of carboxylic and amino acids by liquid membranes: Applicability, process considerations, and potential advantages, *Biotech. and Bioeng.*, **41**, 287 (1993).
- Way, J.D., Nobel, R.D., Flynn, T.M. and Soan, E.D., Liquid membrane transport: A survey, *J. Membrane Sci.*, 12, 239 (1982).
- 8. Wan, Y.H., Wang, X.D. and Zhang, X.J., Treatment of high concentration phenolic wastewater by liquid membrane with N_{503} as mobile carrier, *J. Membrane Sci.*, **135**, 263 (1997).

- 9. Ho, W.S., Hatton, T.A., Lightfoot, E.N. and Li, N.N., Batch extraction with liquid surfactant membranes: A diffusion-controlled model, *AIChE J.*, **28**, 662 (1982).
- 10. Bhowal, A. and Datta, S., Studies on transport mechanism of Cr (VI) extraction from an acidic solution using liquid surfactant membrane, *J. Membrane Sci.*, **118**, 1 (2001).
- 11. Jabbari, A., Esmaeili, M. and Shamsipur, M., Selective transport of mercury as $Hg_2Cl_4^{2-}$ through a bulk liquid membrane using K^+ -dicyclohexyl-18-crown-6 as carrier, *Sep. and Puri. Tech.*, **24**, 139 (2001).
- 12. Zhang, R.H. and Xiao, L., Design of a liquid membrane system for extracting rare earths, *J. Membrane Sci.*, **51**, 249 (1990).
- 13. Ho, W.S.W. and Sirkar, K.K., Membrane Handbook, Chapman & Hall Inc. N.Y., (1992).
- 14. Rautenbach R. and Machhammer, O., Modeling of liquid membrane separation processes, *J. Membrane Sci.*, **36**, 425 (1988).
- 15. Cahn, R.P. and Li, N.N., Separation of phenol from wastewater by the liquid membrane technique, *Sep. Sci.*, **9** (6), 505 (1974).
- 16. Terry, R.R., Li, N.N. and Ho, W.S., Extraction of phenolic compounds and organic acids by liquid membranes, *J. Membrane Sci.*, **10**, 305 (1982).
- 17. Teramoto, M., Takihana, H. and Shibutani, M., Extraction of phenol and cresol by liquid surfactant membrane, *Sep. Sci. Tech.*, **18**, 397 (1983).
- 18. Kakoi, T., Goto, M., Natsukawa, S., Ikemizu, K., Nakashio, F., Matsumoto, M. and Hano, T., Recovery of phenols using liquid surfactant membranes prepared with newly synthesized surfactants, *Sep. Sci. Tech.*, **31**, 107 (1996).
- 19. Correia, P.F.M.M. and Carvalho, J.M.R., Recovery of 2-chlorophenol from aqueous solutions by emulsion liquid membranes: batch experimental studies and modelling, *J. Membrane Sci.*, **179**, 175 (2000).
- 20. Correia, P.F.M.M. and Carvalho, J.M.R., A comparison of models for 2-chlorophenol recovery from aqueous solutions by emulsion

- liquid membranes, *Chem. Eng. Sci.*, **56**, 5317 (2001).
- 21. Zhang, T., Huang, B., Zhou, W., Gao, H., Wu, H. and Wu, J., Extraction and recovery of gold from KAu(CN)₂ using cetyl-trimethylammonium bromide microemulsions, *J. Chem. Tech. Biotech.*, **76**, 1107 (2001).
- 22. Schlosser, S. and Kossaczky, E., Comparison of pertraction through liquid membranes and double liquid-liquid extraction, *J. Membrane Sci.*, **6**, 83 (1980).
- Sastre, A., Madi, A., Cortina, J.L. and Miralles, N., Modeling of mass transfer in facilitated supported liquid membrane transport of gold (III) using phospholene derivatives as carriers, *J. Membrane Sci.*, 139, 57 (1998).
- 24. Fu, J., Nakamura, S. and Akiba, K., Liquid membrane transport of gold by a trioctylamine mobile carrier, *Analytical Sci.*, **10**, 935 (1994).
- 25. Deng, T.L., Liao, M.X., Wang, M.H., Chen, Y.W. and Belzile, N., Enhancement of gold extraction from bio-oxidation residues using an acidic sodium sulphite thiourea system, *Minerals Eng.*, **14**, 263 (2001).
- 26. Yan, N., Shi, Y. and Su, Y., A study of gold extraction by liquid membranes, *Water Treatment*, **5**, 190 (1990).
- 27. Ensafi, A.A. and Eskandari, H., Efficient and selective extraction of iodide through a liquid membrane, *Microchemical J.*, **69**, 45 (2001).
- 28. Mukkolath, A.V., Gaderkar, P.T. and Tiwari, K., A new method for the reduction of swelling in liquid emulsion membrane systems, *Chem. & Ind.*, 192, 19 March (1990).
- 29. Yan, J. and Pal, R., Osmotic-swelling behavior of globules of W/O/W emulsion liquid membranes, *J. Membrane Sci.*, **190**, 79 (2001).
- 30. Walstra, P., Principle of emulsion formation, *Chem. Eng. Sci.*, **48**, 333 (1993).
- 31. Clausse, D., Pezron, I. and Komunjer, L., Stability of W/O and W/O/W emulsions as a result of partial solidification, *Colloids and Surfaces A*, **152**, 23 (1999).
- 32. Bobin, M.F., Michel, V. and Martini, M.C., Study of formulation and stability of emulsions

- with polymeric emulsifiers, *Colloids and Surfaces A*, **152**, 53 (1999).
- 33. Shere, A.J. and Cheung, M., Modeling of leakage in liquid surfactant membrane system, *Chem. Eng. Comm.*, **48**, 143 (1988).
- 34. Trang, Y.S., et al., The use of grey-based Taguchi methods to determine submerged arc welding process parameters in hard facing, *J. Materials Processing Tech.*, **128**, 1 (2002).
- 35. Fisher, R.A., Statistical methods for research workers, Oliver & Boyd, London, (1925).
- 36. Kargari, A., Extraction of gold (III) ions from aqueous solutions using emulsion liquid membrane system, Ph.D. Thesis, Amirkabir University of Technology, Tehran, Iran, (2004).