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The Effect of Type and Concentration of Receiving Phase on Silver Separation Efficiency by Supported Liquid Membrane Technique

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Abstract
The Effect of The Type and Concentration of The Receiving Phase on The Efficiency Of Silver Separation with Liquid Supported Membrane Techniques. The process of making negative x-ray films in a radiology laboratory produces liquid wastes containing various chemical compounds with the main content being Ag metal (in the form of Ag+ cations). This waste is categorized as a dangerous and toxic material. Therefore it is necessary to separate the Ag metal before it is discharged into the environment so as not to be harmful to life and the environment. One of the separation techniques that can be used is with supported fluid membrane technique (Supported Liquid Membrane, SLM). The supported liquid membrane has three important components: the feed phase containing the component to be separated, the membrane phase containing the carrier compound and the receiving phase containing the integral component. The three-phase composition determines the efficiency of the separation process. This study aims to identify the type and concentration of the receiving phase solution to improve silice metal separation efficiency by SLM technique. To obtain an efficient composition in the recipient phase was carried out by varying the type of acid received solution ie H3PO4, HNO3 and CH3COOH with respective concentrations of 0.01; 0.05; 0.10; 0.15; and 0.20 M. Measurement of metal ion concentration of Ag+ before and after transport was determined by Atomic Absorption Spectrophotometer (SSA) at wavelength 328,22 nm. Based on the result of research that type and concentration of acid solution in the receiving phase has an effected on separation efficiency. The optimum transport percentage has obtained on the use of H3PO4 receptor solution at a concentration of 0.1 M with a percentage of Transport Ag of 46.84%.

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Abstrak
Proses pembuatan negatif *film* foto rontgen di laboratorium radiologi menghasilkan limbah cair yang mengandung berbagai senyawa kimia dengan kandungan utama adalah logam Ag (dalam bentuk kation Ag^+). Limbah ini dikategorikan sebagai bahan yang berbahaya dan beracun. Oleh karena itu perlu dilakukan pemisahan logam Ag sebelum dibuang ke lingkungan agar tidak berbahaya bagi kehidupan dan lingkungan. Salah satu teknik pemisahan yang dapat digunakan adalah dengan teknik membran cair berpendukung *(Supported Liquid Membrane, SLM*). Membran cair berpendukung memiliki tiga komponen penting yaitu fasa umpan yang mengandung komponen yang akan dipisahkan, fasa membran yang mengandung senyawa pengemban dan fasa penerima yang mengandung komponen yang telah terpisahkan. Komposisi ke tiga fasa tersebut menentukan efisiensi proses pemisahan. Penelitian ini bertujuan untuk mengidentifikasi jenis dan konsentrasi larutan fasa penerima untuk meningkatkan efisiesi pemisahan logam perak dengan teknik SLM. Untuk mendapatkan komposisi yang efisien di fasa penerima dilakukan dengan *memvariasikan* jenis larutan penerima asam yaitu H_3PO_4, HNO_3 dan CH_3COOH dengan konsentrasi masing-masing 0,01; 0,05; 0,10; 0,15; dan 0,20 M. Pengukuran konsentrasi ion logam Ag^+ sebelum dan sesudah transport ditentukan dengan Spektrofotometer Serapan Atom (SSA) pada panjang gelombang 328,22 nm. Berdasarkan hasil penelitian bahwa jenis dan konsentrasi larutan asam di fasa penerima berpengaruh terhadap efisiensi pemisahan. Persen transport optimum diperoleh pada penggunaan larutan penerima H_3PO_4 pada konsentrasi 0,1 M dengan persen transport Ag sebesar 46.84%.

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1. INTRODUCTION

Silver is a shiny white metal, corrosion resistant and light and a good conductor of electricity among all metals, even copper. In general, silver is found together with Zn, Pb, Co, Ni and Au (ore). Silver is obtained as a result of melting and refining metal from its ore. Apart from being obtained from mineral ores found in nature, silver metal (Ag) is also sourced from waste such as industrial waste, photography waste, X-ray photo waste, and others.

Photo-X-rays are a technique used to image the inside of an organ or cell tissue (tissue) in the human body. Corny film from photo-X-rays contains many chemicals, one of which is a layer of silver (Ag) on the plastic photo film. In general, liquid waste used from photo-X-ray washing contains silver (Ag) concentrations ranging from 2500.00 mg/L

to 6200.00 mg/L. Based on this, this waste is categorized as B3 waste (Hazardous and Toxic Materials) which can damage and disrupt human life and the environment. Therefore, waste processing must be carried out before the waste is disposed of into the environment to minimize the content of dangerous substances in it (Widayanto et al, 2016).

Separating Process of silver metal waste currently plays a very important role in controlling dangerous heavy metal pollution and recovering valuable silver metal from its impurities. Various methods are used to recover silver metal from these wastes, most of which are effective within certain Ag concentration limits. Silver in the form of an anionic thiosulfate complex [Ag(S2O3)2]3- can be separated from the solution by electrolysis, metal replacement, precipitation, ion exchange. The electrolysis and metal replacement methods change the silver thiosulfate complex compound into Ag metal, while the precipitation technique will change the Ag complex into a precipitate form with the addition of precipitating agents such as sodium sulfide, sodium borohydride and sodium dithionite. (Djunaidi et al, 2007).

Solvent extraction is a good and popular method for the separation of metal ions. One technique developed from the solvent extraction method is the liquid membrane technique. The liquid membrane is a water phase that acts as a selective barrier between two water phases that are homogeneous and do not mix with the membrane phase. The two liquid phases separated by a liquid membrane are respectively called the external phase (feed phase) and internal phase (receiver phase). The feed phase contains components to be separated and the receiving phase contains components that have been separated (Fitriya, 2011). The liquid membrane technique is divided into several parts, namely Bulk Liquid Membrane (BLM), Emulsion Liquid Membrane (ELM), and Supported Liquid Membrane (SLM).

One technique that can be used for the separation of metal ions is the supported liquid membrane (SLM) technique. Theoretically supported liquid membrane is one of the membrane-based separation methods developed from solvent extraction techniques, namely by immobilizing the extracting agent (carrier) on a porous polymer membrane (Basir, 2015). The advantages of the supported liquid membrane technique are that it requires little extractant/supporting compound used for extraction, simple operation and low cost. This method has advantages such as it can be applied even if the concentration of dissolved metal ions is low, the process takes place continuously, and uses a small amount of organic solvent (Parhi, 2013). The supported liquid membrane consists of three important components, namely the feed phase containing the components to be separated, the membrane phase containing the carrier compound, and the receiving phase containing the components that have been separated. The process of transporting metal ions in a supported liquid membrane involves an exchange reaction between H+ ions and the metal ions being transported (Basir, 2015). Because carrier compounds generally have H+ ions that can be exchanged, it is best that the receiving solution used be an acid solution for ion transport in the carrier compound to facilitate the exchange reaction of metal ions from the feed phase to the acceptor phase.

There are various types of acid solutions that can be used as receiving solutions, both strong and weak acid solutions. The acid solution must have a large degree of ionization to be able to provide more H+ ions for good metal separation. Apart from that, the concentration of a solution also determines the optimum performance to obtain higher purification of silver metal. Therefore, the performance of the receiving phase is largely determined by the type of acid and its concentration.

Based on the background description above, it is necessary to carry out research to determine the effect of the type of receiving phase solution and its concentration on the efficiency of separating silver metal using the supported liquid membrane technique.

2. RESEARCH METHOD

Tools and materials

The tools used in this research are: a set of distillation tools and chemical glasses, a set of SLM separation tools, an atomic absorption spectrophotometer (SSA) to determine the percent transport of metal ions. Meanwhile, the materials used in this research are: AgNO3 solution as a silver standard and as a solution in the feed phase, di-2 ethylhexylphosphate (D2EHPA): tributylphosphate (TBP) as a carrier compound, H3PO4, HNO3 and CH3COOH as a solution in the receiving phase, folitertrafluoroethylene membrane (PTFE) with a diameter of 47 and a pore size of 0.5μm as a supporting membrane, kerosene as an organic solvent for supporting compounds and distilled water.

Research procedure

The supported PTFE membrane is soaked in a combined carrier compound solution consisting of D2EHPA: TBP (4:1) with a total concentration of 0.5 M then taken and placed in such a way in the SLM tool between the feed phase (silver nitrate solution) and the receiving phase. Separation process Silver ions from the feed phase to the receiving phase through the membrane phase were carried out for 5 hours with a stirring speed of 300 ppm.

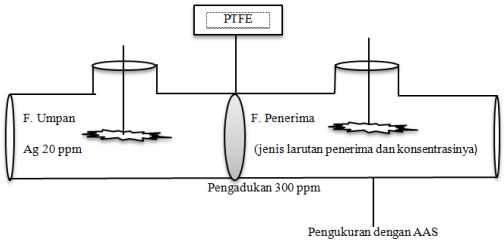


Figure 1. SLM tool

In this experiment, the variables that were made constant were 100 mL of 20 ppm AgNO3 solution in the feed phase, a combined carrier compound solution consisting of D2EHPA: TBP (4:1) with a total concentration of 0.5 M in the membrane phase, while the variables that were made changed were receiving phase with varied types and concentrations, namely 100 mL of HNO3, CH3COOH and H3PO4 solutions with concentrations of 0.01 each; 0.05; 0.1; 0.15 and 0.2 M.

The Ag concentration of the feed phase before the separation process and the Ag concentration in the receiver phase after the separation process were measured using an Atomic Absorption Spectrophotometer (AAS). Separation efficiency is known through high extraction percentage using the formula:

$$\%transpor = \frac{C_x}{C_o} X100\%$$

Where:

C_x= Concentration of Ag transported to the receiving phase

C_o= Initial Ag concentration in the feed phase

3. RESULTS AND DISCUSSION

A. Research result

The effect of the type of receiving phase solution and its concentration on the transport percent of silver metal

To study the effect of the type of receiving phase on the process of re-releasing metal ions from their complex form with carrier compounds in the membrane phase to the receiving phase, this is done by varying the type and concentration of the receiving phase solution. The receiving solutions used in this study were HNO3, CH3COOH and H3PO4 with each concentration varied at 0.01; 0.05; 0.10; 0.15; and 0.20 M. The results

of transport of silver metal from each recipient solution at varying concentrations can be shown in Table 4.2 below:

Table 1. Effect of type and concentration of receiving solution on the percent transport of silver metal

Type of Solution Recipient Acid	Concentration				
	0.01 M	0.05 M	0.1 M	0.15 M	0.2 M
H3PO4	31.60%	45.74%	46.84%	43.47%	38.46%
HNO3	20.68%	35.12%	39.01%	28.56%	33.98%
CH3COOH	14.26%	22.86%	22.37%	22.66%	23.46%

Table 1 shows that the higher the concentration of the receiving solution used, the higher the silver metal transported. Optimum transport is obtained when using a H3PO4 receiving solution with a concentration of 0.1 M, where the silver transport percentage is46.84%.

B. Discussion

The effect of the type of receiving solution on the transport percent of silver metal using the supported liquid membrane technique

The type of receiving solution plays a roleroleimportant for the process of separating metal ions from the phasebaittowards the receiving phase. The presence of an efficient receiving solution will facilitate the release of metal ions that form complexes in the carrier compound. To release silver metal ions from their complex form with the membrane phase to the receiving phase, a receiving solution is needed that is capable of providing H+ ions. The presence of H+ ions in the receiving phase can facilitate the release of metal ions that form the complex into the receiving phase. In this study, the types of receiving solutions used were HNO3, CH3COOH and H3PO4 with respective concentrations of 0.01, 0.05, 0.10, 0.15, and 0.20 M. The results of measuring the transport percent of silver metal using each receiving solution and its concentration can be shown in Figure 2 below:

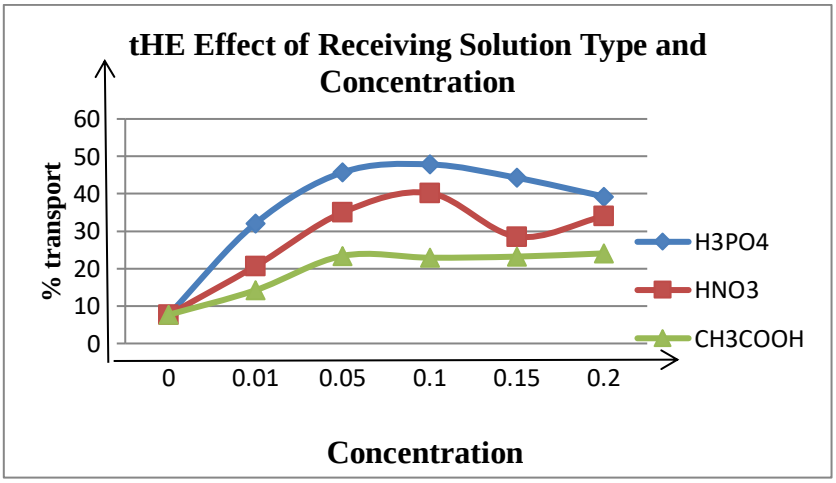


Figure 2. Percent transport of silver metal using each solution and concentration

Figure 2 shows a graph of the relationship between the type of receiving solution and the variation in concentration of each solution to the percent transport of silver metal. Based on Figure 4.3, silver metal transport without the presence of an acid solution in the receiving phase gives a silver metal transport percentage of 7.64%. The presence of various types of acids as receiving solutionsshowthe transport percentage of silver metal is increasing. At the same concentration, a higher transport % was obtained in the H3PO4 receiving solution, namely 46.84% when compared to the HNO3 and CH3COOH receiving solutions. The difference in the percent transport of silver metal is influenced by the structure of each recipient solution. The molecular structure of each recipient solution can be shown in Figure 3 below:

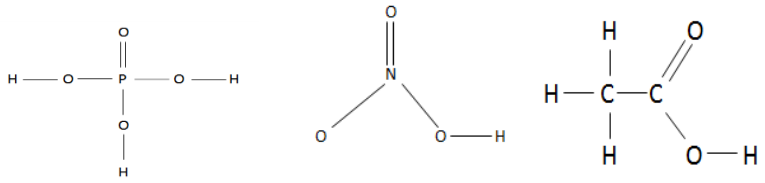
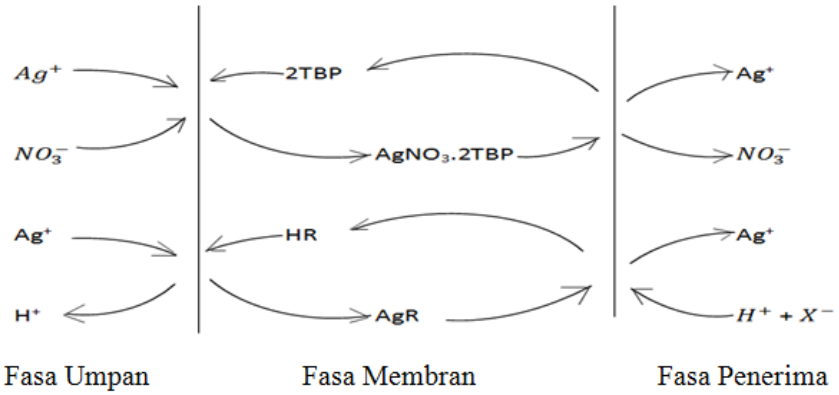


Figure 3. structure of each recipient solution

Figure 3 shows the structure of each recipient solution. The structure of H3PO4 shows that phosphate ions can attract metal ions which form complexes more easily to the receiving phase by forming Ag3PO4 when compared to using an HNO3 receiving solution which only forms AgNO3. Meanwhile, CH3COOH as the receiving solution provided the lowest transport percentage, namely 23.46%. This is because silver metal ions will find it difficult to form the AgCH3COO complex compound in the receiving phase so that silver metal ions which form a complex in the carrier compound will be difficult to release into the receiving phase.

The general transport mechanism for silver metal ions is from the feed phase to the receiver phaseon a supported liquid membrane are as follows:



Judging from this mechanism, it shows that there is an ion exchange reaction from the phasebaitto the receiving phase on the supported liquid membrane. At the interface between the feed phase containing Ag+ metal ions and the membrane phase containing the carrier compound TBP: D2EHPA, an ion exchange mechanism occurs between Ag metal ions and H+ ions in the carrier compound. When the TBP compound forms a dimer, it has two lone pairs of electrons found on the oxygen atom so that it can form coordination compounds with metal ions and can dissolve the complex formed. Metal ions tend to bond to oxygen which has a lone pair of electrons in the TBP molecule. Meanwhile, the compound D2EHPA (RH) is a one-base acid compound, so it can be written as HDEHP (di-ethyl hexyl phosphoric acid). In this situation, the D2EHPA

compound which forms a dimer will tend to break one of its hydrogen bonds to bind the metal ion. The breaking of this bond causes the metal ion to form a complex (AgR) in the carrier compound. The formation of this complex results in an unstable carrier compound. Therefore, at the interface between the membrane phase and the receiving phase, an ion exchange mechanism occurs again between the silver metal ions which form a complex (AgR) in the carrier compound and the H+ ions which are in solution in the receiving phase. The ion exchange mechanism occurs causing silver metal ions to be in the receiving phase and the carrier compound will return to its original form to bind the next metal ion. The pushing force that occurs in this process is due to the potential difference between the feed phase and the receiver phase (Basir, 2015).

The effect of receiving solution concentration on percent silver metal transport

To determine the effect of the concentration of the receiving solution on the percent transport of silver metal, a concentration variation of 0.01 was carried out; 0.05; 0.10; 0.15 and 0.20 M. The percentage value of silver metal ions is calculated from the number of ions transported from the feed phase to the receiver phase through a supported liquid membrane. The calculation results show that variations in the concentration of the receiving solution have an effect on the percent transport of silver metal using the shielded liquid membrane technique as shown in Figure 2.

Figure 2 shows that by increasing the concentration of the receiving solution the percent transport also increases. This indicates that the decomposition speed of the complex in the receiving phase is influenced by the concentration of the receiving phase solution. At higher concentrations, the silver metal ions transported to the receiving phase decrease. This is because the large number of silver ions trapped in the membrane phase which forms a complex will be more stable so that the decomplexization process due to the presence of H+ ions in the receiving phase is reduced (La Harimu et al, 2010).

4. CONCLUSIONAND ADVICE

Conclusion

Based on the results of the research and discussion, it can be concluded that: the type of receiving solution and its concentration influence the efficiency of separating silver metal using the supported liquid membrane technique. The highest transport percent was obtained when using H3PO4 receiving solution at a concentration of 0.1 M with an Ag transport percent of46.84%.

Suggestion

This research is research that studies the factors that influence the transport of silver metal through a supported liquid membrane. To further increase the silver transport percentage, it is necessary to study the influence of other parameters such as the addition of complexing compounds in the receiving phase so that metal ions that are already in the receiving phase do not return to the feed phase.

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