



Emulsion liquid membrane stability in the extraction of ionized nanosilver from wash water



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ABSTRACT

The discharge of ionized nanosilver into environment triggers a great concern owing to the toxicity problem for aquatic organism. In this study, emulsion liquid membrane used to extract the ionized nanosilver from wash water. Variables like carrier, stripping agent and surfactant concentrations, emulsifying time, homogenizer and agitation speed, pH feed phase, and effect of ionic liquid [BMIM]⁺[NTf₂]⁻ were investigated. The membrane phase containing Cyanex 302, Span 80, acidic Thiourea, and kerosene as carrier, surfactant, stripping agent, and diluent respectively. Results demonstrated that 99.89% of silver ion was extracted and ionic liquid show good performance on emulsion stability with 10% swelling.

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1. Introduction

Nanosilver has been extensively used in various consumer products, medicines, and food applications owing to the high prospects of its antimicrobial properties. For instance, this nanoparticle can act as an anti-HIV drug by inhibiting the human immunodeficiency virus (HIV) replication [1]. Besides, they are also coated with the medical devices in order to avoid the occurrence of infection [2]. United States Environmental Protection Agency (USEPA) indicates silver as a priority pollutant in natural waters in 1977. USEPA also regulates a guideline of less than 0.10 ppm of total silver in drinking water due to the potential effects of silver ingestion specifically argyria disease. USEPA, however, prescribes maximum concentrations of 3.2 ppb silver in fresh water and 1.9 ppb in salt water based on acute toxicity of silver to macroinvertebrates and fish [3]. These standards are enforced through the issuance of discharge permits at the state level. With respect to the chronic water quality to protect aquatic life, a few states have set the threshold concentrations for example the North Carolina Division of Water Quality has proposed a criterion of 0.06 ppb [4]. Oregon also has established a 0.12 ppb criterion in its administrative rules based on chronic toxicity to rainbow trout and minnows in fresh water and to mysids in salt

water [5]. Texas, New York, and several regions in California have established maximum contaminant levels for silver between 5 and 10 ppb.

Nanosilver used in some products can enter the environment as individual nanoparticles, small clusters, or potentially dissolve into ions. The ionization behavior of nanosilver in the ionic form has been reported by several researchers via clothing, textiles, washing machine, and home application [6–9]. According to Benn et al. [6], about 1.30 ppm of nanosilver ions have been leached into distilled water from the six types of commercial socks containing nanosilver. Another study also observes as much as 11 ppb of nanosilver ions have been released from washing machine (Samsung brand) which contain ion generating devices for nanosilver ions release in order to protect our clothing from bacteria during the washing process [9]. To date, Benn et al. [10] have found that about 3.2 and 7.6 ppm of silver ion was released from silver nanoparticles coming from the detergent and the toothpaste, respectively. The presence of nanosilver in the natural water environment is of great concern due to its small size, which can create toxicity effect to aquatic organism, especially when it is in the free ionized form [11]. The toxicity effect occurred when the silver ions have massively associate with the other chemicals organic or colloidal, hence influencing the bioavailability and the bioaccumulation of nanoparticle into the cell of aquatic organism. The observation shows that the exposure of nanosilver ions with concentration of 5 ppm can increase the mortality and heart malformation in zebra fish [12].

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Therefore, the removal of this nanoparticle from the waste water is very positive for environmental safety. In the past, several numbers of methods were reported for nanosilver ion extraction including electrowinning [13], electrochemical method [14], solvent extraction [15,16], electrodialysis [17], and liquid membrane separation [18,19]. The disadvantage of electrochemical method is the deposition rate and the composition of the solution, in some cases, can cause the production of dendrites and loose or spongy deposits and interference from the hydrogen evolution reaction. In addition, this process also involves a large capital investment with expensive electricity supply. Besides, electrowinning also is a suitable process for the treatment of effluents with a silver concentration between 2 and 12 ppm. The recovery of silver down to concentrations of less than 0.2 ppm enables the formation of the precipitate silver sulfide on the cathodic surface. However, a second treatment is required by cementation or chemical precipitation. Solvent extraction also involves high consumption of chemical reagents. The electrodialysis is an advance technology, which does not lend itself readily to small-scale applications due to its high capital and operational costs, and requirement for highly trained people with supervision. Liquid membrane technology is regarded as one of the effective method, which combines the extraction and recovery process thus making this technique more advantageous and profitable over other separation methods. Therefore, this method does not need the second treatment like electrowinning, which need the other treatment, which is cementation or chemical precipitation process [13]. The extraction chemistry is basically the same as that found in solvent extraction, but the transport is governed by kinetic rather than equilibrium parameters, that is, it is governed by a non-equilibrium mass transfer. This process also provides low cost and energy because not much chemicals and equipments are used if compared with the electrochemical deposition method, which involve large capital investment with expensive electricity supply [14]. On the other hand, this method also provides the low consumption of chemicals which is the use of carrier is ten times lower than that used in solvent extraction method [15]. Principally, emulsion liquid membrane involves a dispersion of emulsion containing of organic and aqueous stripping phase into the aqueous feed phase containing solutes. The solute from the feed phase permeates into the membrane phase and chemically reacts with the stripping agent present in the stripping phase, which impenetrates in the membrane phase and remains confined in stripping phase. The transportation of solute in the emulsion liquid membrane is chiefly motivated by the concentration gradient and pH. Based on the ELM system, which involves the mass transfer mechanism of metal ions from the external into the internal phase have proven that the silver ion have been ionized from the silver nanoparticles and react with the carrier and stripping agent.

However, the main drawback experienced with this method is the emulsion stability problem which influences the extraction efficiency. The emulsion liquid membrane stability problem which take place in emulsion liquid membrane system are swelling and breakage phenomena. Swelling occurs when the water molecules from the external phase is transferred inside the internal phase, diluting the solute and reducing the solute concentration. Emulsion swelling can increase in emulsion volume during operation and trigger the breakdown of the globules which finally decrease the extraction efficiency. In the meantime, the membrane breakage occurs when portions of the internal phase spill into the external phase. During breakage, the stripping agent and previously extracted solute is leaked into the external phase [20]. The most stable emulsion was obtained when there was neither swelling nor breakage or emulsion with minimum of swelling occurrence.

The application of ionic liquid in the liquid membrane has become the other alternative in order to develop emulsion stability as well as the extraction efficiency. The unique characteristic of ionic liquid such as low melting point, insignificant vapor pressure, and low flammability have successfully drawn a great attention of many researchers in order to improve the membrane stability. Room temperature ionic liquids, which are constituted by salts comprising organic anions and cations have provided great potential application as an alternative candidate in membrane separation. They have been reported by a number of researchers as a carrier, diluent, and stabilizer in liquid membrane procedure for various types of metal removal [21–23]. In the presence of ionic liquid, the robust membrane barrier has been produced in order to stabilize the emulsion in the emulsion liquid membrane process. The emulsion stability is due to the Coulombic interactions of the charges on the ions of ionic liquids $[BMIM]^+[NTf_2]^-$ with carrier, surfactant, diluent, and stripping agent. The Coulomb interaction also called Coulomb force or electrostatic force between two or more charged. If the particles are both positively and negatively charged, the force is repulsive and if they are of opposite charge, it is attractive. The attractive interaction between the opposite charges of ionic liquid can create the strong van der Waals interactions, which can avoid the coalescence of the internal droplets [24]. This ionic liquid also is capable of developing a polymeric structure with large cavities when they are used as a solvent. These polymeric structures of ionic liquid also act as barrier in membrane phase thus enhancing emulsion stability [25]. The properties such as high hydrophobicity of $[BMIM]^+[NTf_2]^-$ also prevent the reaction with water in the stripping phase, hence enhancing the emulsion stability [26].

In this research, the main focus was to study the extraction of ionized nanosilver from the wash water using emulsion liquid membrane process. Several parameters such as effect of carrier, stripping agent and surfactant concentration, emulsifying time, homogenizer and agitation speed, pH of feed phase, extraction time, and treat ratio were investigated. In addition, the effect of ionic liquid $[BMIM]^+[NTf_2]^-$ has also been attempted as well. The ranges of parameters studied are illustrated in Table 1.

2. Extraction mechanism of ionized nanosilver

The mechanism for the extraction of ionized nanosilver is exhibited in Fig. 1. The extractant, Cyanex 302 act as a carrier in this process. The structure of Cyanex 302 is shown in Fig. 2(a). Silver ion chemically reacts with the carrier to form the silver-carrier complexes, $Ag(RH)_2$ on the external interface as illustrated by Eq. (1). Then, these complexes diffuse through the liquid membrane to the internal interface.

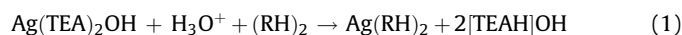


Table 1

The range of parameters used in the extraction of ionized nanosilver.

Operating conditions	Ranges
Volume of acidic Thiourea (ml)	5
Volume of Cyanex 302 in kerosene (ml)	5
Emulsifying time (min)	5, 10, 15, 20
Span 80 concentration (% w/v)	1, 3, 5, 7
Cyanex 302 concentration (M)	0.0003, 0.0005, 0.0007, 0.001
Homogenizer speed (rpm)	8000, 10000, 12000, 13000
Agitation speed (rpm)	200, 250, 300, 350
pH of wash water	2, 4, 5, 8
Thiourea concentration (M)	0.5, 1.0, 1.5, 2.0
Sulfuric acid concentration (M)	1
Ionic liquid concentration (% w/v)	1–6
Treat ratio (emulsion to external ratio)	3, 5, 7, 10
Extraction time (min)	5, 10, 15, 20

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