Supported Liquid Membrane Technology: Advances and Review of its Applications

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ABSTRACT

The present paper gives an overview of the basic principle and mechanism of supported liquid membrane technology (SLMT). Owing to stringent environmental regulations, there is a need for new and unconventional methods of waste purification. The recognition of this technology as a possible solution, along with its wider acceptance in preference to the conventional separation processes, has led to remarkable advances in this field. Salient features of SLMT such as high separation selectivity, simultaneous extraction and stripping, and uphill transport have been briefly discussed. Materials used for different components of the assembly, and innovations in the same, have also been reviewed. Solutions for overcoming setbacks such as relative instability and short lifetime have been assessed. Recent advances and application-oriented research have been reviewed, with the purpose of evaluating their feasibility and effectiveness. Novel processes contrived on SLMs have been proposed as efficient methods for the selective extraction of chemical species in dilute streams, such as metal ions, biological compounds, organic compounds, radioactive species, and gas mixtures. The simultaneous extraction - stripping, mediated by SLM, offers an innovative solution to the dire problem of chemical separation.

Key words: Supported liquid membrane, Waste water treatment, Extraction, Metal ions.

1. INTRODUCTION

Liquid membrane technology has recently garnered attention as a green alternative to separate organic or inorganic substances with minimal energy consumption and high fluxes along with reduced volume of waste. It presents many advantages over conventional separation methods such as adsorption, ion exchange, chemical precipitation, reverse osmosis, solvent extraction processes, and so forth. These include high selectivity and technical feasibility. One of its defining features is that it combines extraction and stripping into one single stage thus resulting in a compact, economical system. The separation is not limited by the conditions of equilibrium since it acts on non-equilibrium mass transfer characteristics.

A liquid membrane is essentially a thin layer of organic phase between two aqueous phases of different compositions. This thin layer of organic phase can be immobilized onto an inert microporous film, and consequently placed in between two aqueous solutions. This arrangement is called supported liquid membrane (SLM). SLMs are used widely over emulsion Liquid membrane and bulk liquid membrane since the membrane thickness required is less, thereby reducing solvent requirements. A thin membrane also ensures high flux and selectivity.

SLMs have found their way into a variety of applications ranging from environmental applications such as heavy metal ion extraction, CO2 capture, and wastewater treatment to difficult and sensitive separations such as acids from aqueous solutions, recovery of drugs, and pharmaceutics. Various application-based advances aim at overcoming some of the drawbacks of this technology, namely, instability of SLMs and its short lifetime. Most research work has been directed at improving these shortcomings by changing the materials used for support, testing new extractants, evaluating green solvents, etc. Several works have proposed new applications for this technology and evaluated their feasibility.

2. MECHANISM

SLMs employ a tri-phase technique to extract solutes from a continuously flowing aqueous stream through a liquid organic phase with subsequent stripping into another aqueous phase. This thin layer of organic phase is immobilized onto an appropriate inert microporous support and interposed between the two aqueous solutions (Figure 1).

Transportation in liquid membranes is a dynamic and non-equilibrium process. The compound that is to be transported has to dissolve in the organic liquid phase and then diffuse to the aqueous stripping phase. This transport takes places due to the difference in the chemical potential across the membrane.

The following processes are involved in the transport of species from feed solution to strip side:
1. Diffusion of substrate across the boundary layer of the feed phase.
2. Extraction of species from feed to membrane on the interface.
3. Diffusion across boundary layer of membrane on feed side.
4. Convective transport across the liquid membrane layer bulk.
5. Diffusion across boundary layer of membrane on strip side.
6. Extraction of species from membrane to strip phase on the interface.

7. Diffusion of substrate across the boundary layer of the strip phase (Figure 2).

The overall membrane flux during this process is determined as follows:

$$J = k_m \Delta C / d$$

Here, km is the mass-transfer coefficient and is proportional to $K_m D_m$, where,

K_m is the partition coefficient,

D_m is the diffusion coefficient,

ΔC is the concentration difference between the strip and feed phases,

d is the membrane thickness.

There are two types of transfer phenomenon of a substance through the membrane phase can be accomplished that is by passive transport or facilitated transport (Figure 3).
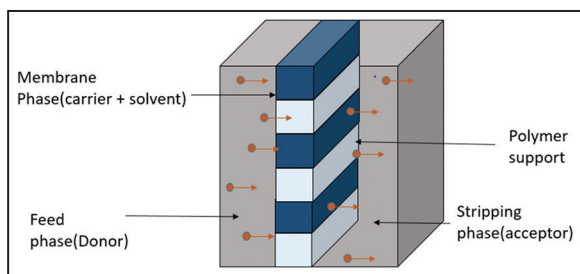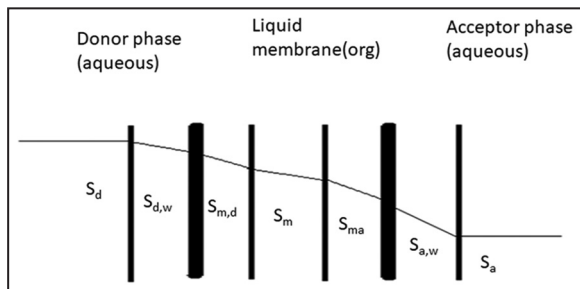

Figure 1: Operation of a supported liquid membrane [1].

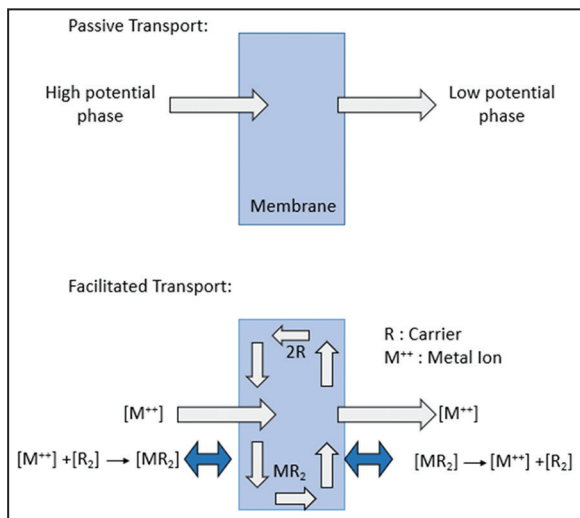**Figure 2:** Transport of a species "S" across a supported liquid membrane.

Figure 3: Passive and facilitated transport.

In passive transport, the substrate dissolves in the organic membrane phase and diffuses to the strip phase due to the concentration gradient between the two phases. On the other hand, in facilitated transport, the membrane phase contains a carrier dissolved in an organic solvent. Since the carrier helps in promoting the transfer rate by increasing the solubility of the permeant, it acts as a catalyst to the process.

3. MATERIALS

SLMs generally consist of three essential components: A polymeric support, a carrier and an organic solvent (or diluent). Performance parameters such as permeation rate and separation efficiency depend strongly on the type of liquids and supports used for SLM construction.

3.1. Support

The support is generally a porous solid membrane (polymer or ceramic) which holds the liquid membrane phase. Thermal and chemical stability toward the feed and receiving phases is a prerequisite for the support material. High hydrophobicity, high porosity, small pore size, and high tortuosity are desired characteristics of a support.

Commonly used supports for flat sheet and hollow-fiber configurations are polymers whose solute flux lies in microfiltration range, such as polyethylene, PTFE, polypropylene, polysulphones, among others. Composite membranes are also available, where the active membrane material is laminated to a coarse material [2] (Table 1).

3.2. Extractive Agent

The extractive agent or carrier is an amphiphilic molecule that ensures and promotes the transport of a substrate from the source phase to the receiving phase. It can be described as a complexing agent or ligand that associates with the substrate to be transported or extracting. It is chosen based on the selectivity of the components present in the feed phase. This molecule should be strongly hydrophobic to minimize its loss by dissolution in the aqueous membrane phases and stay trapped in the organic membrane phase. However, this agent must have a hydrophilic part for binding to the substrate. The most conventional carriers [7] are carboxylic acids, phosphoric, sulfonic acids, amines, and crown ethers used to extract the metal ions or organic compounds ions (Table 2).

3.3. Solvents

A solvent or diluent can be used to prepare various concentrations of the extractants. Some commonly used diluents are kerosene, xylene, toluene, hexane, cyclohexane, and dodecane. One of the challenges in the membrane formulation is to lower the solvent viscosity for better diffusivity of the solute complex within the membrane while minimizing solvent loss (Table 3).

4. STABILITY OF SLM

The most fundamental setback associated with the SLMT is its stability. It is the ability of membranes to operate for an extended period without compromising the quality of separation.

In SLM, the membrane solvent is held merely by capillary forces in the pore structure. During operation, some amount of solvent may be washed or forced out of the membrane under the influence of lateral shear forces. The liquid membrane phase is gradually lost in the aqueous phase at the interface of the membrane on both sides by emulsion formation. Depending on the amounts of solvent and carrier lost from pores, the solute flux decreases, increases, or does not change. The cases are illustrated in Figure 4.

Large pore sizes of the support, wettability of support by aqueous phase, interfacial tension between membrane and aqueous phase, and properties

Table 1: Supports used in SLM configurations.

Type	Process	Properties	Applications
Polysulfone membrane graphene nanopowder was incorporated into PES dope polymer solution to fabricate flat sheet PES membrane support [3]	Prepared by phase inversion process in which polysulfone is mixed with a solvent to form a solution from which the membrane is cast [2]	Increased hydrophobicity of the grapheme-doped PES membrane (contact angle increased to 120° as compared to previous 85°). The force required to break the was higher compared to the pristine PES membrane (4.3N vs. 1.9N). [3]	Biological extraction, separation of acid-water mixtures, extraction of drug molecules
Polypropylene membranes (Accurel and Celgard)	Manufactured by a multistage process including extrusion, annealing, and stretching of isotactic polypropylene to induce the formation of elongated pores. Also produced by thermal phase inversion process	SLMs with the thicker Accurel support show higher stability as a support compared to the thinner Celgard membranes [4]. This is because a larger thickness eliminates water bridging between feed and strip phases through membrane pores	Application for the recovery of heavy metals in hydrometallurgical processes [2]
PTFE fluoropore FG is an asymmetric membrane which one microporous membrane (e.g., PTFE) is used for immobilization of organic phase and the second, thicker porous membrane is used as support	The membranes are made by a patented process in which an impervious PTFE film is converted to an expanded and mechanically stronger film [2]	This film consists of a number of small nodules interconnected by a mass of very fine fibrils. The pore size of this kind membrane can be varied by careful process control. Provides the contrasting properties of high flux and well as mechanical stability	Acid–water mixture separation, alcohol–water separation
PIMs are a type of liquid membranes composed of a liquid phase and a base polymer	PIMs are formed by casting a solution containing a carrier (extractant), a plasticizer, and a base polymer, such as CTA or PVC, to form a thin, flexible, and stable film [5]	The polymer forms a gel-like network that entraps the carrier and plasticizer/modifier. PIMs are also characterized by higher mechanical strength and stability in comparison with other SLMs [6]	Separation of metal ionic species such as Cu, Ni, Pb, Ce, Co.

SLM: Supported liquid membrane, CTA: Cellulose triacetate, PVC: Polyvinyl chloride, PIM: Polymer inclusion membrane, PES: Polyethersulfone

Table 2: Extractants used in SLMT.

Extractant	Application	Comments
Chelating or acidic extractants	Used for the extraction of various metal ions such as Cu, Zn, Co, Ni, Fe, Mn, Mo (VI)	-
Amine-based extractants Alamine-336, Aliquat-336, Alamine-304, etc.	Used for extraction of Mo, Cr, V, and so forth from their chloride solutions.	-
Crown ethers	Used for the extraction of alkali or alkaline earth metals	Can be designed to show selectivity for one alkali metal over another. But these crown ethers are not used in SLM because of their high cost and high water solubility

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of the carrier are some factors that influence emulsion formation. Thus, they play an important role in affecting membrane stability.

Yang *et al*. [17] introduced a P84 copolyimide with novel chemical cross-linking modification which increased the hydrophobicity of both surfaces and demonstrated that it can be effectively used as the polymeric microporous matrix as a viable and economic method to improve the stability of SLM. It was observed that SLM with a symmetric support matrix was comparatively much more stable than that with an asymmetric matrix. This is because in the symmetric matrix, the forces exerted will be balanced both at interfaces such that the liquid membrane phase is not lost from either sides.

Ho *et al*. developed new technique of SLMs with strip dispersion. In this technique, a constant supply of membrane phase is maintained to the SLM to ensure prolonged stability. The technology provides operational stability, is efficient and easy to operate. Applications of SLM with strip dispersion have shown substantial growth in the last 10 years. However, these have been primarily concerned with recovery and removal of metals [18].

Kumar *et al*. [19] experimentally investigated the prevalent reasons for instability and proposed solutions by measuring performance of SLM for phenol extraction. The study infers that stability is enhanced by choosing solvent with high viscosity, adding an electrolyte in the aqueous solution

Table 3: Commonly used solvents in SLMT.

Solvent/diluent	Application	Comments
RTILs	Selective separation of organic compounds and gas separation [8-11]	Possess high ionic conductivity, thermal stability, and non-flammability. Their high surface tension, viscosity, and non-volatility can prevent solvent loss from the porous membrane
"Green liquid membranes"[12] can be prepared using vegetable oils	Soybean oil-based bulk liquid membrane for Cu (II) removal and recovery from aqueous solutions [13]	Non-toxic, environment-friendly. Can be cost-effective as well.
DESs are a new generation of solvents, in which two or three components are associated with each other through hydrogen bond interactions	Recently, CuCl/DESs-SLMs have proved to be a promising method for the separation of olefin-paraffin mixtures [14-16]	Compared to ionic liquids, DESs have several more advantages, such as biocompatibility, biodegradability, easy preparation and availability, low cost, and facile scale-up

RTILs: Room temperature ionic liquids, SLMT: Supported liquid membrane technology, DES: Deep eutectic solvents
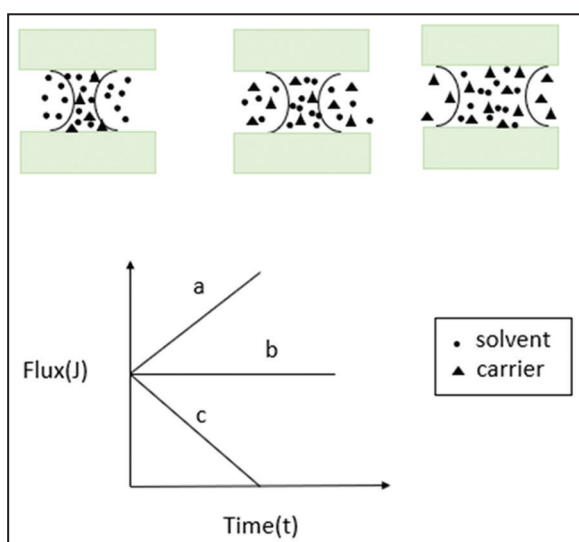

Figure 4: Degradation of supported liquid membrane and its influence on flux (J) during leakage of (a) organic solvent, (b) both organic solvent and carrier, (c) carrier.

so as to increase the interfacial tension between liquid membrane and aqueous phase increasing the stability up to 120 h. Surfactant addition in the liquid membrane arrests the loss of membrane from the pores enhancing the stability up to 144 h. Furthermore, applying a polymeric gel layer on the outer surface of SLM reduces emulsion formation at the interface by hindering contact between aqueous and organic phase. This results in higher stability up to 168 h at the cost of decreased recovery of phenol. The polymeric layer provides additional resistance to mass transfer. Hence, it is important to assess all the parameters affecting stability and flux and optimize the system such that desired flux can be obtained at significant stability of the liquid membrane.

Sun *et al*. investigated how modification in hydrophobicity of support can be used to enhance the stability of SLM. This was done by grafting heptadecafluoro-1,1,2,2-tetradecyltrimethoxysilane (FAS) and SiO2 on the conventional polypropylene hollow-fiber support. It resulted in significant enhancement of stability and an average removal percentage of phenol from coal gasification wastewater (CGW) as feed was maintained up to 75.32% during 16 days of experimentation at pilot scale level [20]. Hence, the system can be potentially scaled-up for large-scale applications such as removal of phenols from industrial wastewater.

Leon *et al*. [21] developed a novel method for SLM preparation which is ultrasound assisted. The method was tested for cobalt (II) removal from aqueous solution. On comparison with SLMs prepared by soaking polymeric material in the carrier's organic solution under atmospheric pressure and in vacuum, it was shown that the former method resulted in higher transport efficiency (>5%) and stability (>6% in the second run and 10% in the third run) of the membrane. This was explained by effects of acoustical streaming and cavitation on the support structure which increased its porosity and pore density. It also led to an increase in pore filling due to decreased viscosity on account of temperature increase caused by cavitation. Although this technology is successful in enhancing the flux, transport efficiency, and stability of the membrane, its commercialization is limited by drawbacks such as control of membrane erosion and poor development of transducer technology. In some cases, ultrasound irradiation can damage the membrane surface [22].

Use of ionic liquids in SLMs to form supported ionic liquid membranes (SILMs) is an emerging technology which has been used to prepare stable membranes. Ionic liquids possess characteristics such as high viscosity, adjustable hydrophobicity and polarity, negligible vapor pressure, selectivity toward gases, and high surface tension which enable them to produce mechanically stable SLMs that are more tolerant to mechanical forces, temperature, and pressure changes. Negligible vapor pressure minimizes their tendency to be soluble in surrounding phases. High viscosity results in greater capillary forces which reduce the displacement of the liquid from micropores due to pressure [23]. Use of expensive ionic liquids is feasible for formation of SILMs since a relatively lesser quantity of liquid is required, unlike solvent extraction. The characteristics of high surface tension and undetectable vapor pressure ensure no evaporative, dispersive, or dissolution loss of membrane. Wang *et al*. conducted separation of toluene/cyclohexane mixtures using SILMs for a period as long as 550 h with a separation factor in the range of 15–25. The membrane showed satisfying durability in permeation of vapor. In addition, strong interactions between the RTIL and the substrate improve the stability of the membrane [24].

SILMs can be exploited for separation of organic/aqueous or organic/organic mixtures with promising results. This technology can also be potentially applied to remove volatile organic compounds from industrial emissions.

The operational stability of SILM is influenced by the ionic liquid immobilization method used for membrane preparation. When immobilization is carried out using vacuum, less amount of highly viscous ionic liquid is absorbed in the pores and results in higher

losses of liquid during operation. Alternatively, the pressure technique is suitable for immobilization of highly viscous ionic liquids. It ensures that higher percentage of membrane pores are filled with the liquid [25].

Treatment with CF4, N2, and O2 cold plasma on SILMs showed an improvement in retention of ionic liquid in the membrane and hence slight increase in its stability [26]. Cold plasma treatment by CF4 increases the hydrophobic nature of the support, whereas treatment by N2 or O2 increases its hydrophilic nature. When the RTIL phase and support are of the same nature (hydrophobic/hydrophilic), the retention of RTILs inside the support will increase. The plasma treatment can be chosen based on nature of impregnated liquid. Such approach of surface treatment can optimize the stability of SILMs on industrial scale. Cold plasma treatment on the surface would be an expensive alternative and hence further studies need to be conducted in developing economically viable surface modification techniques.

5. RECENT ADVANCES AND APPLICATIONS OF SLM IN SEPARATION TECHNOLOGY

5.1. Metal Extraction

Industrialization has led to an exponential growth in the consumption of metals in the past few decades. The commercial importance of these metals combined with environmental problems consolidates the need for recovery of these metals.

Metal ions such as lead and cadmium are some of the undesirable species found in contaminated wastewater. The extraction and transport of cadmium (II) and lead (II) in SLM using organophosphoric compounds such as tributyl phosphate, tris-ethylhexyl phosphate, trioctylphosphine oxide (TOPO) and di-(2-ethlyhexyl)phosphoric acid as mobile carriers have been studied by Azzoug et al. [27]. This SLM system studied could potentially be used for purification, separation, and removal of cadmium and lead from their mixtures after its validation in terms of reproducibility and membrane stability. Advantages of TOPO are that it is immiscible in water, has high basic strength, and compatible with several solvents to give high extraction efficiency. However, organophosphate compounds are a common cause of poisoning worldwide and hence their proper handling and phase contamination needs to be taken care of, thereby adding to the cost [28].

Conventional hazardous solvents have been used in most of the studies on SLMs. In contrast, Bhatluri et al. [29] have utilized a coconut oil-based flat-sheet SLM to investigate the simultaneous extraction and recovery of lead and cadmium from aqueous solutions. Coconut oil, being eco-friendly and inexpensive, was the prime appeal of this technique. The resultant SLM comprising of coconut oil as solvent was also found to be quite stable (up to 98 h) with regard to flux. The optimum extraction and recovery of Pb were found as 84% and 78%, respectively, and that of Cd were found as 79% and 67%, respectively, in simultaneous transportation. These figures are much higher than the extraction efficiencies attained by the previous systems [27]. Moreover, relatively less concentration (0.5%v/v) of carrier (Aliquat 336) was required for efficient transportation of metal ions as opposed to similar feed concentrations reported by other researchers for the same purpose.

Ni and Co have similar chemical and physical properties. Furthermore, their high purity is required in most applications. This has led to the development of unconventional separation routes. SLMs using carrier amines have been used for the selective extraction of cobalt from acidic media, constituting mixtures of cobalt and nickel. Synergistic effect on flux was observed by the inclusion of a modifier, tributyl phosphate (TBP) in the membrane phase. However, optimum conditions required concentration of TBP as high as 25%w/w [30].

The process allowed optimization by varying pH, temperature, carrier concentration, complexing reagent concentration, diluent, etc. Leon et al. conducted studies for Ni-Co separation using SLMs that were prepared by ultrasound-assisted methods and showed that these SLMs showed higher separation factors (almost 20 fold higher) than reported in the literature, in which SLMs prepared by classical methods. This can be attributed to the fact that ultrasound-assisted preparation results in increased porosity and higher pore size of the polymeric support leading to greater extent of pore filling [31]. Thus, the increase in cost due to ultrasound-assisted preparation needs to be weighed in with the increase in separation factor.

The uneven distribution of rare earth metals (REMs) and rare metals in nature and the regulations for their export from country's producers have caused gradual rise in their prices. REMs also play a vital role in the advancement of green energy. Hence, it is vital to devise an efficient REM recovery process from discarded REM containing products. One such metal is Indium. The demand for indium can be met by reusing and recycling indium from electronic wastes. SLMs have not been employed commercially since they are still plagued with instability. SLM with the additional modification of strip dispersion has been a recent development to solve this issue. A hollow-fiber SLM (HFSLM) with strip dispersion system has proved to be successful in the recovery of indium from discarded LCD [32]. Similar technology was also used for extraction of Dy(III) [33], fumaric acid [34] cephalexin [35], etc.

An SLM system, that uses polymeric hollow fiber modules, has also been developed for selective extraction and recovery of REMs such as Nd, Pr, and Dy without coextraction of non-REMs from permanent neodymium-based magnets with neutral extractants like tetraoctyl digylcol amide (TODGA). The distribution coefficients (D) of Dy and Nd extracted by Cyanex 923 ranged from 0 to 19, while that by TODGA were >100 at both 3M HNO3 and HCl [36].

The SLM system operates under non-equilibrium conditions to minimize coextraction of non-REEs [37]. In traditional solvent extraction, separation occurs due to the differences in Ds at equilibrium, which can limit the selectivity of REEs over non-REEs. This may explain the presence of small amounts of Fe and B in the strip solution on extraction with TODGA under equilibrium conditions.

5.2. Radioactive Metals

The nuclear waste that stems from the reprocessing of spent fuels is termed as the high active waste which on concentration yields high-level waste (HLW) and is an abounding source of several radioactive elements with half-lives up to millions of years. Safe and apposite management of radioactive waste is essential taking into account the potential hazards of radioactive elements to human health and environment. The conventional technologies employed for radioactive waste processing have several drawbacks such as high energy consumption and secondary waste formation. SLMT is considered a feasible alternative which could not only supercede these setbacks but also complement these methods while upgrading the parameters of discharges and purification economy. In most of the studies, nitrate media was used to match the acidity of the HLW which is equivalent to 3–4 M HNO3.

The separation of thorium and uranium ions is of some interest and importance. Extraction of uranium ions from nitrate media was demonstrated [38] using a hollow fiber liquid membrane employing TBP-based extractant. Hollow fiber membrane module provides large extraction surface per unit volume of membrane module meaning smaller footprint. However, only up to 67% of uranium ions were extracted while rejecting thorium ions in the raffinate phase. Extraction of uranium ions from acidic medium was also been studied using PTFE SLMs, wherein up to 80% of uranium ions were extracted [39,40].

Recovery of uranium by SLMT has been reported from several waste streams using extractants as diverse as TBP, D2EHPA, bis(2,4,4 trimethyl pentyl)phosphinic acid (Cyanex 272), 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester (PC88A), and amines [41-46].

Although the long-term menaces of HLWs are due to the long-lived radionuclides, for a relatively shorter time period, the chief concern is due to the heat-emitting radionuclides such as ^{137}Cs and [90]Sr. Cesium recovery from alkaline/acidic waste streams has always been a challenge.

Recovery of Cs and Sr has also been demonstrated using crown ethers as carriers [47-49]. Crown ethers although desirable carriers on account of their inert nature are expensive to synthesize and have high water solubility. This has resulted in the advent of several relatively economical amide-based carriers.

Rathore *et al*. [50] have applied the SLM technique using hollow fiber configuration on process scale for the recovery of radio-cesium, which resulted in highly selective recovery of Cs (I) over other radiotracers with good decontamination factors. A mixture of 0.1 M TODGA (tetraoctyldiglycolamide) + 0.5 M DHOA (N, N-dihexyloctanamide) was observed to be a suitable carrier for maximum transport of the metal. The high decontamination factors, as well as high throughputs, encourage the possible application of the system for the recovery of radiocesium from HLW.

Some of the applications of SLM in extraction of metals and radioactive metals have been tabulated (Table 4).

5.3. Wastewater Treatment

Uncontrolled release of non-biodegradable and toxic species into the ecosystem through the discharge of industrial process streams is a major concern since these species get further accumulated in living beings through water consumption. It is therefore critical to extract these toxic species before discharging them into the environment. SLMT emerges as one of the most viable alternatives for this purpose. To mitigate uncontrolled waste of these pollutants, it is essential to develop techniques that permit their recapture and recycle.

Chromium concentration >0.25 mg L^{-1} poses serious threats to not only aquatic but also to human life. Selective and efficient extraction of Cr (VI) from wastewater through SLM using TOPO as a carrier has been reported. When this SLM system was applied on paint industry wastewater, about 80% of chromium (VI) was positively removed from the wastewater [53]. The extraction efficiency can be further increased by employing multiple stages or increasing concentration of carrier in the membrane phase.

Recovery of chromium (VI) using SLM with TBP as carrier has also been studied and the developed system was successfully applied to the effluents from the plating industries [54]. Extraction of Cr (VI) has also been demonstrated employing carriers such as quaternary ammonium salts [55] and amines [56]. Quaternary ammonium salts are environmental-friendly and capable of providing an inexpensive way of extraction.

Ali *et al*. achieved selective removal of zinc (II) from aqueous solutions using a flat-sheet polypropylene-SLM system with triethanolamine dissolved in cyclohexanone as the carrier for extraction. Extraction of zinc ions with 87% of extraction efficiency was reported [57]. The system has been successfully employed for zinc extraction from dry battery wastewater.

The removal of pyridine from aqueous solutions using HFSLM has been studied [58] which confirmed that HFSLM technology is better over the conventional separation technology due to negligible losses

and less extractant consumption.

Due to the high content of phenols in CGW, the wastewater cannot be degraded with the help of traditional biological methods directly, implying that the removal of phenols is a key step before the biological treatment process. Removal of phenols from the CGW employing a polypropylene HFSLM has been achieved [59,60].

However, high organic concentration in CGW at times may degrade stability of the membrane by accelerating dissolution of the liquid membrane phase. Hence, selection of appropriate SLM has to be followed by careful assessment of constituents of the wastewater. The work of Msagati *et al*. [61] successfully employed the SLM extraction technique for monitoring trace amounts of ionizable organic contaminants, which include 17b-estradiol and its metabolites, benzimidazole anthelmintic antibiotics, testosterones, and their methyl ester derivatives, sulfonamides in wastewaters.

Recovery and separation of nickel [62,63], copper [64,65], zinc [66], precious metals [67,68], rare earth metals [69], alkali metals [70], and so forth, from industrial wastewaters using SLM have been extensively studied and vast experimental surveys been conducted.

5.4. CO₂ Capture

It is essential to develop novel processes for separating the primary greenhouse gas component, carbon dioxide from gas streams. Membrane gas separation provides higher energy efficiency, lower cost, and makes the process environmentally benign and flexible compared to conventional absorption methods used for separation. A drawback of using this process is that there is seldom a good balance between ideal selectivity and permeability through solid membranes. SLMs help to overcome these limitations.

A study by Cheng *et al*. [71] provides a potential method for sequestering carbon dioxide directly from the atmosphere using microporous SILMs preventing the need to extensively transport CO2 to the industries requiring it from units where specialized absorption systems that capture CO2 are setup. Out of the symmetric and asymmetric microporous polyvinylidene difluoride (PVDF) membranes fabricated, the symmetric SILMs were shown to be capable of effectively separating low concentration of CO2 from ambient air due to high CO2/air selectivity. This method is environment-friendly, cost-effective, and consumes lesser energy compared to conventional absorption/desorption methods.

Protic ionic liquids (PILs) show high selectivity and a reversible reaction with CO2. Hence, their incorporation in SLMs may lead to better selectivity and permeability of for CO2 separation. Zhang *et al*. [72] introduced PILs to prepare SILMs and investigated the system for selective separation of CO2/CH4 and CO2/N2 gas pairs. Four diamine-monocarboxylate-based PILs were used in this work and were found to be selective and tunable carriers of CO2 in the presence of water. Furthermore, they exhibited higher permselectivity and competitive permeability as compared to supported PIL membranes at ambient temperatures. Further research has also been carried out to evaluate the separation efficiency of CO2/CH4 and CO2/N2 using (3-aminopropyl) trimethoxysilane and acetic acid and which showed very high selectivity of CO2/CH4 [73]. Administration of PILs in SLMs is still in its development stage and additional research is required to assess its reproducibility, scale-up, and stability.

A new class of SILMs consists of task-specific ionic liquids [74]. In amino acid ionic liquids, the amino group can react with CO2 and hence work as CO2 carrier. Tetrabutylphosphonium glycine ([P(C4)4] [Gly] has very high CO2 absorption capacity in both, humid and dry conditions in contrast to general SILMs which have lower CO2

Table 4: Different SLMs employed for various metals and radioactive metals.

Metal extracted	Polymeric support	Solvent	Extractant	Ion flux/mass transfer performance	Ref
Co	Microporous hydrophobic polypropylene Celgard 2500	Chloroform	Alamine 308	Permeation coefficient at pH 4 is $1.30*10^{-5}$ m/s	[30]
Cd (II) and Pb (II)	Celgard 2400	Chloroform	D2EHPA, TOPO	Permeability of Pb (II) at 0.01M D_2EHPA is $5*10-0.5$ cm/s Permeability of Cd (II) at 0.01M D2EHPA is $3*10^{-0.5}$ cm/s	[27]
Indium	Polypropylene HFSLM with strip dispersion system	Kerosene	D2EHPA	Distribution ratio of indium at pH = 2 is 20	[31]
REMs (Nd, Pr, Dy)	Polypropylene HFSLM	Isoparafin	Tetraoctyl diglycolamide (TODGA), Cyanex 923	Distribution coefficient of Nd extracted = 21 by TODGA in 2M HNO_3	[36]
Hg (II), Cd (II), and Cr (III)	PVDF membrane	dodecane	Isooctylmethylimidazolium bis-2-ethylhexylphosphate	extraction of Cd (II) ions at pH = 3 (higher permeability coefficient: $40.3 \times 10-7$ cm^2 s^{-1}	[51]
U, Th	Celgard microporous polyethylene fibers wrapped around central tube feeder and shell casing in polypropylene HFSLM	kerosene	TBP	Permeability = 0.009 cm/s for 500 ppm concentration of uranium ions in feed phase with containing 1 M HNO_3	[36]
Am (III)	Supported PTFE membrane	IDA/n-dodecane	(N, N,N',N' -tetra-2-ethylhexyl) 7-oxabicyclo[2.2.1]heptane-2,3-dicarboxamide (OBDA)	diffusion coefficient value of $5.1 \times 10-6$ cm^2/s	[52]
Cs (I)	Polypropylene HFSLM	NPOE and n-dodecane	calix-[4]-bis (2, 3-naptho)-crown-6 as the mobile carrier	Diffusion coefficient = 2.2×10^{-7} cm^2/s	[47]
Sr (II)	Polytetrafluoroethylene	a mixture of 2-nitrophenyl ether and n-dodecane	di-tert-butyl-dicyclohexano-18-crown-6 (DTBCH18C6)	Permeability = $2*10^{-04}$ cm/s	[48]
Cr (VI)	Celgard 2400	Toluene	TOPO	Flux of $1.4*10^{-7}$ mol/cm^{2}s	[53]
Zn (II)	Flat-sheet polypropylene SLM	Cyclohexanone	Triethanolamine (TEA)	Flux of $3.5*10^{-11}$ mol/m^{2}s at 50%TEA	[32]

NPOE: 2-nitrophenyl octyl ether, HFSLM: Hollow fiber supported liquid membrane, TBP: Tributyl phosphate, IDA: Isodecyl alcohol

selectivity in the presence of water. Another study that used SLM consisting of amine terminated ionic liquid for separation of CO2 from CO2/CH4 mixture exhibited high selectivity and stability (>260 days) [75]. Although task-specific ionic liquids have excellent selectivity toward CO2, they demonstrate increase in viscosity on contact with CO2 which leads to reduction in flux.

Yahia *et al*. [76] developed a thermally stable biomimetic membrane system that employed SILMs and an extreme-enzyme to enhance the selective transport of carbon dioxide at high temperatures. It is expected that such systems will bridge the gap between thermal stability of enzymes and enhanced membrane performance at elevated temperatures for separation of CO2. The transport selectivity toward CO2 against N2 was reported to be quite high with separation factor of CO2/N2=35.6. These results show that this approach can be further extended for removing CO2 from flue gases by enhancing the performance of enzyme impregnated SILMs. Such an inclusion of enzymes in SLM is an illustration of scope of enhancement of carrier activity to increase the flux.

5.5. Extraction of Carboxylic Acids

Carboxylic acids are among the most important organic pollutants discharged from industrial sources. Sources of these carboxylic acids include manufacture of cellulose acetate, chemical pulping of wood, and the waste from petrochemical industries. Carboxylic acids results in the increase of oxygen demand when released in water.

From a practical point of view, it is important not only to extract carboxylic acid but also to concentrate it.

It has been shown that carboxylic acids can be efficiently extracted from diluent aqueous streams [77] using SLM technique. TOPO has been shown to be an effective extractant for extraction of acetic acid, formic acid, and phenylacetic acid. TOPO is immiscible in water, has a high basic strength and compatible with several diluents in giving relatively high extraction efficiencies. Moreover, scale-up experimentation was conducted for this system and same flux was obtained at both scales. Results scaled with interfacial area.

Recovery of fumaric acid was achieved by HFSLM with strip dispersion system using trialkylamine carrier. The stability of the HFSLM with strip dispersion system was remarkably good with no significant losses of solvent from the membrane [77]. Therefore, the SLM system can be considered as a newly developed method to separate and recover fumaric acid from waste effluents. The present type of HFSLM with strip dispersion system cannot entirely avoid the pollution of the aqueous solutions with membrane solution due to its dissolution in both aqueous strip and feed solutions. Further research needs to be conducted to solve this issue, using membrane solution with water-insoluble components.

SLM has also been employed in separation of citric and lactic acids from aqueous solutions [78], extraction of citric acid from fruit juices [79], recovery of acetic acid using vegetable oils [80], among many others.

5.6. Biological Advances

SLMT has found its way in bioanalysis for analyzing samples of human plasma, urine, serum, and saliva. Extractions of specific components from these samples are carried out using SLM.

Blood proteins like human serum albumin interfere with capillary electrophoresis (CE) analysis on direct injection of blood samples and hence their effective elimination is of prime importance. Kubáň *et al*. [81] developed a simplistic sample cleanup device comprising of planar SLM integrated to CE for the direct injection of human body fluids. The analytical procedure was demonstrated by online injection, rapid pretreatment, and CE determination of certain amino acids in plasma samples and human serum. This method holds and advantage over traditional methods of analysis of human body fluids as it provides an online solution which leads to a fast, effective, and efficient process. Previous attempts to create an online process were fraught with problems such as necessity of external instrumentation, low reproducibility, elaborateness, and possible sample carryover which were successfully overcome with this novel system.

A method for the determination of ketamine and its primary metabolites such as norketamine and dehydronorketamine in urine samples was investigated [82] using hollow-fiber liquid phase microextraction (HF-LPME) in three-phase mode. This method proved to be better than conventional liquid extraction since very high analyte enrichments could be obtained due to the high sample-to-acceptor volume ratio. In addition, by employing essential eucalyptus oil as the liquid membrane a green approach was achieved. This type of extraction could eliminate the use of hazardous organic solvents in conventional sample preparation methods based on HF-LPME.

The content of saliva is a perfect reflection of the composition of blood, however, at significantly reduced concentration. This makes saliva not only an excellent candidate for a clinical analysis but also requires a highly sensitive analytical technique. A way around this is analyte enrichment in the sample for easy detection. Tudorache *et al*. [83] monitored progesterone in saliva with the help of magnetic particle-based immune-SLM assay in a sequential injection setup that allows analyte enrichment, automatic sample cleanup, and detection in a single analysis unit. However, the validation of this method in terms of selectivity and robustness remains and there is scope for miniaturization of this system. [81,85,86]

The efficient transport of selected peptides through SLM comprising cationic carriers was depicted by high extraction efficiency [84]. In these cases, the stability of SLM was observed to be high (about 2 months) using Aliquat 336 as a carrier. However, for extraction of peptides from blood plasma, the system with Aliquat 336 encountered setbacks such as adsorption of plasma proteins on the membrane surface, which limited the mass transfer of peptides. Therefore, searching for a more suitable method, a new way of peptide extraction using SLM with anionic carriers such as D2EHPA was proposed [85-87].

5.7. Extraction of Pharmaceutics and Drugs

SLMT has been proved to be feasible in recovery of drugs and pharmaceutics such as salbutamol, terbutaline, and cephalexine [87,88]. The problem of extraction of hydrophilic drugs such as salbutamol and tertbutaline was overcome by the use of cationic carries like Aliquat 336.

Balchen *et al*. [89] demonstrated extraction of acidic drugs across a thin hydrophobic SLM by the application of an electrical potential difference. It is a rapid process with equilibrium and requires organic solvent volume in microliters to sustain the SLM. The nature of the SLM was observed to be highly influential, and long-chain alcohols with strong proton acceptor properties were found to be suitable. This process allows for fine tuning and optimization by varying the pH conditions in the donor and acceptor solutions and the voltage for the electric field.

Gjelstad *et al*. [90] demonstrated tuning of the liquid membrane to selectively extract polar or non-polar drugs. The liquid membranes composed of 2-nitrophenyl octyl ether (NPOE), di-(2-ethylhexyl) phosphate (DEHP), and tris-(2-ethylhexyl) phosphate (TEHP) as extractants in the study of electrokinetic migration of different drugs. Pure NPOE was efficient in extracting non-polar drugs, whereas extraction of polar substances required addition of DEHP to the liquid membrane. Combining NPOE, DEHP, and TEHP resulted in a liquid membrane for simultaneous extraction of both non-polar and polar drug substances. Hence, electrokinetic transport across liquid membranes may be altered dramatically by the composition of the liquid membrane, which may help to effectively control selectivity.

6. CONCLUSION

SLM is one of the most promising and potential technologies leading to its several applications. The wide range of SLM applications show that it is an attractive choice when a selective and efficient method is necessary. Owing to its several advantages such as high selectivity, low energy consumption, no phase contamination, ease of operation, and low cost operation factors, it is being reflected that SLMT would provide a suitable and efficient alternative to the existing conventional separation processes. It uses neither pressure nor voltage, rather is based on difference of chemical potential as a driving force of the process. In the last 10 years, extensive research has been conducted to increase the applicability of SLMT in the metal separation and recovery, pharmaceutical and chemical industry, gas separation, wastewater treatment, biotechnology, and analytical chemistry. Despite, SLMs have not been employed commercially, primarily because of their major setback, instability. Out of the number of studies proposed on improving stability of SLM such as gelation, hydrophobicity enhancement, surface coating, among others, strip dispersion technology is believed to provide a promising solution to stability constraints of SLM on industrial scale. On the basis of the numerous advantages offered, there is no doubt that they possess immense commercial potential. One can expect SLM technology to procure its rightful place in a range of commercial applications in the near future.

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