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Transport of Metal Ions and Small Organic Compounds using Liquid Membranes and Polymer Inclusion Membranes: A Review

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Abstract: ***Extensive work in the field of solvent extraction, liquid membrane transport and transport through PIM has been reported in the recent years and seldom on SLM, ELMs and composite membrane. Out of the available method of separation PIM studies have gained popularity as technological use has been made possible for large scale separation and hence their application in environmental science and hydrometallurgy are obvious. PIM &BLM have demonstrated great degree of selectivity for numerous anions and also small organic molecules. They have the advantage that extraction & de-extraction can be achieved in single run. This review provides detailed overview of all such researchers that have been carried out in past six years and for comparison finding of some earlier years.***

Keywords: transport, liquid membrane, polymer inclusion membrane, carrier, polymer, plasticizer

Abbreviations

ACM, Activate composite membrane; AFM, Atomic force microscopy; (β-CD), β-cyclodextrin; BLM,Bulk liquid membrane; CTA, Cellulose triacetate; Cyanex 302, Bis(2,4,4-trimethylpentyl)monothiophosphinic acid; Cyanex 471X, Tri-isobutylphosphine sulfide; CYPHOS IL 101, Trihexyl(tetradecyl)phosphonium chloride; CYPHOS IL 102, Trihexyl(tetradecyl)phosphonium bromide; CYPHOSIL104, Trihexyl(tetradecyl)phosphonium-(2,4,4-trimethylpentyl)phosphinate;DBE, dibenzyl ether;DVB, divinylbenzene; D2EHPA, Di(2-ethylhexyl) phosphoric acid; ELM, Emulsion liquid membrane; FTIR, Fourier transform infrared spectroscopy; IL,Ionic liquid; MB, Methylene blue; NPOE, Nitrophenyl octyl ether; NPPE, 2-Nitrophenyl pentyl ether; PIM, Polymer inclusion membrane; PVC, Poly(vinyl) chloride; PVA,Poly(vinyl-alcohol); (PC18AAm), poly(N-oleylacrylamide); SEM, Scanning electron microscopy;SIR,Solvent impregnated resin; SLM, Supported liquid membrane; TBP, Tri-n-butylphosphate; TGA, Thermogravimetric analysis; TIMM, Transmission infrared mapping microscopy;TIOA,Tri-isooctylamine;TOA,Tri-n-octylamine; TOPO, Tri-n-octylphosphine oxide; XRD, X-ray diffraction;

1. Introduction

Liquid membrane concept has been known for over 50 years &it's found quite superior over conventional method of separation such as solvent extraction. This review is an extension of 2006 & 2012 review article of S.D.Kolev wherein focus investigation was PIM. However in this review of ours focal theme is also BLM & SLM along with PIM. Research article on BLM &PIM appear regularly in various journals & focus on the period 2006 to 2015 is emphasized. All these modal membrane transport methods have been targeted for various alkali & alkaline earth metal ions (Sourabh Muktibodh 2015 [13]), transition metal ions, lanthanides &small organic compounds also including some dye stuffs.

In all these method of membrane transport, it is shown that the rate is not classical exponential variation but carrier facilitated one and therefore, the nature of carrier is also very important. Selection of carrier & correct membrane composition offer a very effective method of separation important from commercial point of view, resolving environmental issues and achieving selectivity of high degree. This has been found useful in metal ion recovery in hydrometallurgy, biotechnology & treatment of industrial waste. From chemistry point of view PIM is useful in making ion selective electrodes though, this idea has not been employed commercially as yet. Owing to problems of stability & sensing of polymeric membranes. BLM offers low interfacial surface area & mass transfer rates, while for SLM, stability is a prime concern. PIM is therefore superior technique of metal ion transport over BLM & SLM. However research contribution in BLM & SLM is also significant. Wide range of carriers/ extractants have been employed, particularly macrocyclic crown ethers, acidic phospho - derivatives (Omar Arous et al. 2011 [35]) & commercial Aliquats (Alex Upitis et.al.2009 [38]). This reviews aims at providing the summary of current knowledge relevant to BLM &PIM studies between the years 2006-2016.

PIM technique has been found to be useful in preparation of gold nano particles with controlled size when EDTA as a reducing agent is chosen for PVC containing Aliquat 336 in 1-dodecanal solvent YaYa N Bonggotgetsakul et al. 2011 [55]. Meanwhile SLM despite of their weaknesses (limitation) have been used for carrier facilitated transport of various ions using selected extractants (such as T.Eljaddi et al.2014 [22]). Sufficient mathematical ground is also incorporated in their work.

Recent work on activated composite membrane for the transport ion Ag (I) from aqueous solution offers a new dimension to the membrane transport experiments. This is in the form of Activated Composite Membrane (ACMs) which

immobilized the carrier in one of the polymeric layer that constitute them. Such membranes have higher stability than liquid membrane with comparable metal ion permeation while maintaining all the advantages belonging to liquid membranes. Though the effectiveness has yet to be seen. ACM was obtained by polysulphone casting solution by dissolving in DMF and their deposition on non –woven paper support was immersed in water and carrier was coated with interfacial polymerization (YucundoMwndozaTolenlino et al. 2014 [18]).Polymeric pseudo liquid membrane from poly(N-oleylacrylamide) is reported by Hiroko Shino et.al 2014 [20].This is another way of carrying out membrane transport where in authors have reported transport using various crowns. Typically used for separation of chiral substances from racemic mixtures. In two steps process a PC18AAm was prepared and characterized by GPC followed by this pseudo liquid membrane were prepared by pouring polymer carrier solvent over PTFE membrane filter. The cation analysis was made by conductometric methods. An interesting work of PIM an ionic liquid is reported by Francisca-Tomus-alanso et.al 2013 [24] were dynamic potential response of ions selective electrode is studied. The membranes were prepared by casting PVC and ionic liquids 1-butyl-3-methyl ylidazoniumhexachlorophosphate and trihexyltetradecylphosphonium chloride. Response of membrane towards Ibuprofen organic anions was noticeable and reproducible

2. Transport Phenomenon

Membrane separation is relatively new potential field and has become a centre of large scale industrial activity these days. Micro-filtration, ultra-filtration, dialysis, osmosis etc. are some obvious applications of the field. Parallel to this biological membrane provide a barrier between an intracellular and extra-cellular aqueous environment. These membranes are bilayers of amphiphilic phospholipids and glycolipids which support the membrane proteins that can induce selective transport through the cell membrane.

3. Bulk Liquid Membrane (BLM)

A brief review on BLM (by Jayshree Rajkumar et.al 2015 [9] is also available that demonstrates the continuous interest of researcher in the field. These include separation or extraction of U(IV)from phosphoric acid medium by BLM containing D2EHPA in transport of copper ions.

Transport of alkali and alkaline earth metal ions by selective linear Oxocrown ethers across BLM has been published by authors of the article and it has been demonstrated that the chain length of the carrier, lipophilicity of counter anion, hydration energy of transported metal ion or solvent - philicity of the metal ion plays a crucial role in the metal ion transport. It is suggested that the cation is wrapped by linear chain if Oxo-crown containing oxygen atoms. (Sourabh MuktiBodh 2015 [10])

Similar work by Varsha Gautam & co-workers (2015) is reported using various organic membrane containing dichloromethane, 1,2 dichloroethane and $CHCl_3$ and linear ligands such as tripropyleneglycol monobutyl ethers is reported. The same fact comes out that metal ion transport depends upon lipophilocity of counter ion, dielectric constant of membrane solvent and chain length of carrier. Of course, lipophilicity of counter ion is also important. After all, it is the ion-pair that gets transported to maintain the electrical neutrality. Diffusive transport of selected drugs acetaminophens transport through BLM is reported by Ioana Diacoune 2015 [10]. They have developed mathematical model expressing flux of in presence of carrier.

$$J_{as} = \frac{[L]_m \, D_{Al} \, K_{ex} \, [A]^0_W}{d(1+K_{ex} \, [A]^0_W)}$$

Where,
D_{Al} - diffusion coefficient of complex;
$[A]^0_W$ -Initial concentration of the neutral compound in the source phase;
L_m - carrier concentration in aqueous phase in the membrane;
K_{ex} - is the ratio of compressed membrane conc. in the membrane and product of concentration of ligand and free solute in aqueous phase;
D- dielectric constant;

Optimum concentration for the transport of acetaminophen is established as 10^{-4} mole/ L TBP 10^{-2} mole/ L in $CHCl_3$ 1 mole/ L NaOH in receiving phase.

In an interesting experiment of transport of Cu(II), membrane material used were fresh cooking oil, waste cooking oil and kerosene oil with the extractant D2EHPA and TBP as modifier. Reportedly, transport kinetics of Cu(II) across these membrane constituents was affected significantly, identical papers 2014, 2013 Siu Hua chang et.al. Transport phenomenon using $CHCl_3$ membrane containing synthesized diaza-bibracchial lariat ethers 1-6 is reported by T. S. Ramljak et.al 2012 [28].

The results obtained by extraction experiments showed that the bibracchial lariat ether 4,5,6 in which linkage of the adamantane side - arm to the diaza-18-crown-6- is realized by an amide bond, exhibited practically no extraction capabilities towards any of the alkali metal cation studied. However, lariat ether 2 and 3, in which the adamantane side arm is linked to the diaza 18 crown -6 by an amine bond, showed enhanced extraction pronounced compared to the parent crown ethers 7 and 8 as well as significantly higher selectively of K(I) than diaza- crown. Conformational analysis suggested that the difference in cation binding capabilities of investigated compound is mostly caused by the difference in their conformational flexibility. The result obtain from the transport experiment showed that the transport rates of lariat ethers 2 and 3 for K(I) are larger than for the N, N ,1 - dibenzodiaza- 18 crown -6(8), but significantly lower than for diaza-18- crown-6(7) on the other hand, the difference in the transport male of K(I) vs Na(I) picrates, for both 2 and 3, are pronouncedly larger when compared to the referent crown ethers 7 and 8, what makes these compounds suitable for selective transport of K_j. In similar work Gholamereza et.al. 2012 [29]. Reported that highly selective transport of Pb(II) can be achieved using 1,4,8,11- tetra aza cyclo tetra decane as carrier in BLM transport process. Source phase contained 5 ml of 1.5 x 10^{-4}

M ligand and 1. 0 x 10 $^{-3}$ M oleic acid while the receiving phase contained 10 ml of 0. 1 M $Na_2P_2O_7$. It is demonstrated that ligand is itself is not quite effective in transporting Pb(II) ions but addition of long chain fatty acid such as oleic acid facilitates transport effectively. It is suggested that Nitrogen atom of ligand and proton donor site of fatty acids possibly forms some bonds which imparts a greater degree of lipophilocity to the crown ether metal ion complex to facilitate cation transport through BLM.

Some other noticeable finding has been reported by Niculina - Nina BADEA et.al. 2011[34], G. H. Rounaghi et.al 2009 [37].

4. Polymer Inclusion Membrane (PIM)

Beata Pospiech et .al 2015[4] reported separation of Cd(II) and Cu(II) from their aqueous solution using phosphonium ion liquids in PIM as carriers. CYPHOS IL 101, CYPHOS IL 104 were used in a CTA membrane using ONPOE as plasticizers. In their studies of solvent extraction Cd(II) &Cu(II) from HCl solution were selectively extracted. The extraction efficiency of Cu(II) increases with decreasing pH. Their mechanism is based upon on chloro complexes of these species. Stripping efficiency in sulphuric acid strongly attracts Cd(II) and therefore sulphuric acid is suitable stripping agent in this case. Similarly, transport pattern examine here in CTA membrane shows greater flux for Cd(II) with a selective coefficient of 18.5 and 28.1 in CYPHOS IL 101 & CYPHOS IL 104 respectively. The membrane constituents here are 15.6% CTA 41.2 w% CYPHOS IL 101 & CYPHOS IL 104 and 43.2 w% of ONPOE in enabled extraction above 86% of cadmium. The role of NaCl in source phase adversely affects the transport as demonstrated here.

In another publication of the same author Cyanex 471X carrier in PIM showed effective transport of Pd(II)ions. Membrane of CTA ONPOE and Cyanex 471X was studied were both the aqueous phases were pumped with peristayltic pump. It was shown that carrier concentration plays a crucial role. The initial flux of palladium was only 4.7µ mole.m2sec^{-1} at 0.5 mole dm^{-3} concentration. The highest efficiency for Pd(II)was obtained 2.0 mole dm^{-3} on volume of plasticizers. Thus, an optimum value is observed and it was shown that permeability coefficient also decreased with increasing hydrochloric acid concentration. Effect of KSCN in the receiving phase indicated that at higher molar concentration of KCNS (.5M) initial flux is higher which in turn indicated the recovery factor to be higher. Similar work with CYPHOS IL 101, 102,104 i.e. phosphonium ions liquids for the removal of Pb(II) from aqueous chloride solution in PIM and liquid -liquid extraction process is reported by Rosocka et .al 2015[56].CYPHOS IL 102 is used for the first time as metal ion carrier and is reportedly excellent extracants of Pd(II) ions in both method of extraction i.e. PIM liquid-liquid extraction. About 84-90% of extraction is achieved.

The work O.Kebiche-Senhandji et.al 2015 [6] was focused on extraction recovery of Pb(II) ion by D2EHPA in PIM experiment in CTA membrane 2-NPOE as plasticizer . PIM Morphology was established by SEM and FTIR results were used to conclude the intermolecular interactions between base polymer and extracants. Transport efficiency was found to be about 92% in dilute aqueous solution. However, it was observed that selectivity depends upon relative composition of the membrane.

In other interesting work, modeling of the extraction process for the removal of phenol from aqueous bu Fadila OUGHLIS- Hammache et.al 2015[11]. PVA/β-CD Membrane was developed in a relative weight ratio of 2%-20% weight %. The inclusion complex of phenol in β-CD was confirmed by UV-visible Spectrophotometer. An extraction yield of 33% was obtained after an equilibrium time of 50 minutes. The model revealed the existence of strong interaction between the concentration of solution and stirring phase. In an attempt to explain the extraction mechanism, the theory of “free volume” as proposed by Peng et.al (2006) was used.

Imidazole and its alkyl derivatives have been shown to have excellent extracting abilities for various transition metals ions in the review paper of Spas D.Kolev et. al 2006, 2012([60], [61]); Review of extraction and transport of organic molecules using PIM by Michelle O’Rourke et. al 2009[36] is useful in the sense that it specifically focuses on the transport of the organic molecule such as lower sugars, small acid molecule, lower amino acids, phenol and salicylic acid derivative by various author in the respective years. The work of Malgorzata Ulewicz et. al 2015[5] on PIM experiments doped with l-hexyl-4-methyl imidazole for pertraction of Zn(II) and other transition metal ions from the chloride solution demonstrated that solution membrane has a high flux for Zn(II) ions and acknowledgeable flux in mixtures of Zn(II) Cd(II), Zn(II) Cd(II) Co(II) and Zn(II) Cd(II) Co(II) Ni(II). The selectivity coefficient of Zn(II) ion in all the system is obtained to be 9.4, 9.8, and 12.9 respectively. It was established that initial fluxes for Zn(II) ions in case l-hexyl-4-methyl imidazole as ion carrier is twice as high as that for membrane doped with l-hexyl imidazole. The porosity (ϵ) and roughness (Rq) of the PIM was studied by AFM Diffusion coefficient were calculated in this report and were found to be in the range of $10^{-10} – 10^{-12}$ cm^2 /sec. it was also established that PIM of these carrier is thermally stable compare to l-hexyl imidazole. Similar work has also been reported in their publication in 2011 [33].

The work of Elzbieta Radzy minska- Lenarcik et. al 2015[12] for the separation of Co(II), Ni(II), Cu(II), Zn(II) in PIM interestingly demonstrated the use of steric effects on carrier molecule used in earlier reports series of alkyl imidazole derivatives are used in a CTA membrane ONPOE plasticizers. AFM characterization indicated that the roughness decreases with the branching while the porosity increases. The author claims that a decrease in membrane porosity is due to increase in molar volume of the carriers when substituted. As usual flux increases with carrier concentration and it was observed that Zn(II) and Co(II) ions form complexes with coordination number 4 and 6 and are more easily recoverable with the use of substituted carrier. The series of carriers used are l-heptyl imidazole, l-heptyl-2-methyl imidazole and l-heptyl-4-methyl imidazole.

In the work of Uqur Ayes et. al 2015[39] the transport of Zn(II) ion from aqueous nitrate solution of different metal ions through CTA and 2-NPOE membrane and calix[4] Resorcinarene derivatives was investigated. Membrane characterizations were done by SEM & AFM. Various factors were examined and were compared to relative to SLM experiments. The higher permeability coefficient values of Zn(II) ions were found for SLM compared to PIM. However, Zn(II) more effectively transported in these PIM experiments.

In a recent work of B.N.Mahanty et. al Nov.2015[41] PIM studies were extended to actinide ion separation containing several substituted Diglycolamides. Typical ligands used were TPBGA, THDGA and TDDGA membrane was made by CTA & 2-NPOE. The optimized membrane composition is reportedly is 68.4% and substituted DGAs 17.9% NPOE and 13.7% CTA. PIM Morphology is studied by Transmission infrared mapping microscopy TDDGA was found to be non-homogenous extracants and reached out during the studies. Transport parameters were investigated and effect of alkyl chain length on the uptake of Actinide ions was evaluated. Similarly, work of Xiaorong Meng et. al Nov (2015[40] describes transport of phenol through PIM containing N-N-di (1-methyl heptyl) acetamide as carrier is reported. Thermodymanic parameters suggested that diffusion in the membrane is rate controlled step. The maximum flux was as high 25 mg m^{-2} s^{-1} .The overall kinetics is reportedly first order.

The authors of these articles have developed a PIM containing non-cyclic Oxo-crown ethers as carrier for effective transport of alkali & alkaline earth metal ions. It was demonstrated that a larger cation is transported to a greater extent by a larger carrier. It was also found that lipophilicity of the counter anion is much significant. Transport is higher when picrate is counter anion and least when Ortho-nitro Phenolate is counter anion. The entire process appears to be carrier facilitated one Sourabh Muktibodh 2015[13].

The work of Monika Baczynska et. al 2014[14] on a mixed polymer membrane with ONPOE as a plasticizers and phosphonium ionic liquids CYPHOS IL 101 & 104 showed effective removal of Zn(II) from aqueous chloride solution. In a peristaltic pump experiment (2% CTA 4% PVC 10% ONPOE and .1 M of the carrier), isolated experiments on CTA & PVC membrane were also done. However, it was demonstrated that CTA membrane is performing better over PVC membrane. The maximum transport of Zn (II) is obtained for 40% of carrier concentration in both the cases.
In a different study of PIM transport of some organic acid is demonstrated (such as adipic acid. succinic acid, oxalic acid). In a PIM experiments containing 1- alkoxy methyl imidazole as a carrier. The polymer matrix contained 60% & 40% mass ratio of carrier and polymer with no plasticizer added. In an attempt to describe the flux of organic acids across PIM mathematical model developed is expressed as

$$J = \frac{dn}{Adt} = \frac{V}{A}\frac{dc}{dt} = P.(Co - 2.C) \qquad (1)$$

Where, n-number of molecules transported through the membrane,mol;
t-time,S;
C- Concentration of organic acid in the receiving phase at time t,mol/m^3;
C_o intial concentration of organic acid in the feeding phase,mol/m^3 .
Equation 2 obtained by integration between limits from t=0-t, C=0 a convenient, relationship obtained is -

$$\frac{1}{2}\ln\left(1 - \frac{2C}{Co}\right) = \frac{A}{V}.P.t \qquad (2)$$

The above equation gives the value of slope

$$K = \frac{A}{V} * P$$

and knowing K permeability coefficient P is m/s is determined.

$$P = \frac{V}{A} * K$$

The entire explanation is based upon certain assumption such as (1) reaction rate is high on surface of membrane (2) concentration of transferred compound in the membrane compare to the carrier concentration (so that concentration of free carrier in the membrane is constant) (3) Ficks law is obeyed (4) the diffusion of compound in liquid film is much faster than the diffusion of the complex through membrane and (5) Both the phase are stirred perfectly.

In the experiment it was shown that increasing alkyl chain length of the carrier causes increase in the oxalic acid transport and the overall experiments the highest Flux and permeability coefficient are for the oxalic acid. It was concluded in the experiment that the transport rate of organic acid through PIM decreases with increase carbon chain length between carboxylic groups in the following range

Oxalic acid > Malonic acid > Succinic acid ≳ Adipic acid.

Remarkably, it was established that 1- alkyloxy methyl imidazoles perform a dual role of a plasticizer and carrier of examined organic acids. In the parallel work of transport of organic acids describe above by different but similar carrier is published by Piotr Gajewski et.al 2014[19].

Similar experiments were recorded alkyl chain length of a-11 carbon atoms facilities effective transport and Flux of acids is similar to that mentioned above. Work of Ounissa Senhadji-kebiche et.al 2014[15] in solid phase extraction for water treatment processed describes use of PIM as competitive material. Acidic extrcants like Cyanex 302 was used for removal of Zn(II), Cu(II), Fe(II) ions by PIM & SIR (solvent impregnated resins) in 0.1 M nitric acid solution. Various parameters have been studied in these experiments.

Alamine 336 and TBP as carriers the competitive transport of metal ions shows that preferential selectively order is Co(II) > Cd(II) and thus the method is effective for separation and recovery of Co(II) and Cd(II) from aqueous solutions (Yasemin yildiz et.al. 2014[16]). Here CTA membrane, 2 NPPE and NPOE as plasticizer are used. The average thickness of membrane is estimated to be 25µm. Membrane characterization is done by AFM, FTIR & SEM. In this work of Marta koldziejska demonstrates that N- (diethyl thio phosphoryl) - aza [18] crown -6- could be regarded as an effective agent used for separation of Ag(I) from aqueous solution of nitric acid. The crown other as an ion carrier for

competitive transport of Ag(I) and Cu(II) give preferential selectively order: Ag(II)>>Cu(II) also as ionic extracant gives very high removal for silver cotton with good selectivity and surprisingly the entire amount of Ag(I) can be removed from source aqueous phase under pH 3.0.

The influence of type of polymer matrix on the transport of certain organic acids using PIM is demonstrated. A comparison of 3 polymer matrix CTA, CAP, CAB and 1-alkyl imidazole as carrier with the alkyl chain length of 10,11,12,14 and 16 carbon atoms with the similar theoretical consideration explain earlier become obvious in his work. This research suggests that with increasing percentage of acetyl group in the composition of polymer matrix increases the rate of transport of organic acids CTA happen to be best polymer followed by CAB. CAB is selectively good for transport of tartaric acid. This however is not supported by any theoretical arguments. Hydro metallurgical recovery of Co(II) from acid chloride solution and Cyanex 301 carrier in another PIM experiment of CTA ONPOE. Beata Pospiech 2013[25] proves to be effective in separation of Co(II) from lithium ions. The optimum condition observed are 32 wt.% of Cyanex 301 50 wt % of o-NPOE and 18 wt % of CTA in 3M HCl solution as receiving phase. Recovery of Co(II) is as much as 98% after 12 hrs.

It is reported that life time of PIM can be extend by addition of 0.1 M $NaNO_3$. The reports suggest that life time of membrane can be as high as 52 days by addition of 0.1 M $NaNO_3$ in contrast to the membrane with no $NaNO_2$ where the expected life time about 7 days. This article calculates membrane liquid loss in the transport of phenol by co-poly (Eugenol DVB) as carrier. It is routine PVC membrane where DBE is used as plasticizer. PIM morphology was studied at regular intervals and effect of plasticizer was also investigate

The authors in their earlier publication in (2012) have claimed that the above describe membrane works well for the transport of phenol at alkaline pH. Other factors including membrane thickness have also been shown but again logical arguments are missing L. Saidi mansouri et.al 2012[32] has investigated that morphology and stability of PIM of composition CTA polymer, 2- NPOE plasticizer and $BMIMPF_6$ as carrier (1-N-butyl-3 methyl imidazolium hexafluoro phosphate).

Investigation focused on FTIR analysis Suggest that there is no chemical reaction between PIM constituents (which is of course not a secret) and there are only weak interaction. In an interesting work of cationic dye removal (methylene blue MB) using PIM of Aitali Salima and co-workers 2012[30] phosphoric acid derivative as carrier (D2EHPA) were used. The PIM is composed of CTA, 2-NPOE and D2EHPA. Membrane morphology was characterized by SEM FTIR & TGA methods. Optimized pH and other values have shown that extraction efficiency is as high as 93%.

Similar work appeared in (2012) where the membrane composition and solution chemistry are investigated using PIM containing Aliquat 336 carrier for transport of Cd(II) selectively. Sandra Adelung et.al. 2012[27] The PVC membrane of such kind is used for Cd(II) Cu(II) & Zn(II) transport under different stripping solvents. Negative charge metal chloride complexes were formed as it is claimed. These research is also support by thermodynamic modelling of chloro metal complexes and therefore the role of chloride ion is crucial for both source & receiving phase the essence of these publication is that Cd(II), Cu(II), Zn(II) selectively can be controlled by regulating the chloride concentration in the extraction solution. De- extraction in the stripping phase was favourable in acidic condition. Similar kind of work is also reported by B.Gajda et.al 2012[26]. However it uses different carriers 1- decylimidazole. The work was aimed for the removal of heavy metal ions Ni(II), Co(II), Cd(II), Zn(II) from waste solutions again this was chloride ion controlled transport process. PIM is constituted by CTA, and 1-decylimidazole about 88% of transport efficiency for Zn(II) ion is observed. However no clear-cut application of such a device is indicated.

Table 1

Reference No.	Membrane base polymer	Plasticizer	Carrier	Target metal ions/ molecule	Morphology	Remarks
1	PVC	DEHP	Pyrazole substituted benzene	NH_4(I)	-	2016
2	CTA	NPOE	Hexachlorinated cobalt dicarbollide	Cs(I)	-	2016
3	CTA	NPOE	Dicyclohexan-18-crown-6	K(I)	SEM	2016
6	CTA	NPOE	D2EHPA	Pb(II),Zn(II) &Cd(II)	SEM and FTIR	2015
12	CTA	NPPE	1-Heptyl imidazole, 1-heptyl 2-methyl imidazole, 1-heptyl 4-methyl imidazole	Cu(II), Zn(II),Ni(II), Co(II), selectivity for Cu(II)	Non-contact AFM	2015
11	PVA, extraction process	none	βΔX–	Phenol	XRD, SEM	2015
7	CTA	NPOE	CYPHOS IL 101, CYPHOS IL 104	Cu(II), Cd(II),	-	2015
39	CTA	NPOE	Calix[4] Resorcinarene derivative	Zn(II)	SEM ,AFM	2015
41	CTA	NPOE	Substituted DGAs	Am(III),Pu(IV),UO_2(II), Th(II)	TIMM	2015
40	PVC		N,N-di (1-methylheptyl)acetamide (N503)	Phenol	SEM	2015
56	CTA		CYPHOSIL101 ,102 and	Pd(II)	-	2015

			CYPHOSIL104			
4	CTA	NPOE	Cyanex 471X	Pd(II)	-	2015
5	CTA	NPPE	1-hexayl-4-methylimidazole	Zn(II),Cd(II),Co(II), Ni(II)	AFM	2015
46	PIM		1-alkyl imidazole &TOA	Citric acid	-	2014
14	CTA/ PVC	NPOE	CYPHOSIL101&CYPHOSIL104	Zn(II)	-	2014
	CTA	1-(alkyloxy)methylimidazole	1-(alkyloxy)methylimidazole	Adipic acid,succinic acid .malonic acid,oxalic acid	-	2014
15	CTA/SIR	NPOE	Cyanex 302	Zn(II), Cu(II),Fe(II)		2014
16	CTA	NPOE,NPPE	Alamine336 & TBP	Cu(II),Co(II),Ni(II),Cd(II)	AFM,SEM, FTIR	2014
19	CTA	1-alkyl-1,2,4triazoles	1-alkyl-1,2,4triazoles	Oxalic acid,succinicacid,adipicacid ,malonic acid	-	2014
21	CTA	NPOE, NPPE	TIOA	Co(II),Li(II)	-	2014
	CTA	NPOE	N(diethylthiophosphoryl)-aza[18] crown-6	Ag(I),Cu(II)	-	2014
42	CTA	NPOE	Calix[4]resorcinarene	Zn(II),Cd(II)	FTIR,SEM,AFM	2014
43	CTA		O-phenyl acetyl β-CD	Pb(II),Cd(II),Na(I),Cu(II)	-	2014
44	PIM		TOPO, Aliquat336	Zn(II),Cu(II)	-	2014
45	CTA		Hexachlorinated derivative of CobltBisdicarbollide	Cs(I)	-	2014
26	CTA, PVC		Aliquat336	As(V)	XPS (don't show difference in surface of CTA & PVC)	2014
	CTA,CAB, CAP		1-alkyl imidazole with alkyl chain length of 10,11,12,14&16 carbon atom	Oxalic acid,tartric acid ,lactic acid	-	2013
47	PIM		Calix[4]arene	Cr(VI)	FTIR,AFM	2013
49	PVC		D2EHPA	Ur(VI)	-	2013
	PVC	DBE	Co-poly (eugenol DVB)	Phenol	SEM	2013
25	CTA	NPOE,TEHP, DEHA	Cynex301	Co(II),Li(I)	-	2013
31	PVC	DBE	Co-poly (eugenol DVB)	Phenol	SEM,FTIR	2012
32	CTA	NPOE	BMIMPF6	Cr(VI)	FTIR	2012
30	CTA	NPOE	D2EHPA	Methylene blue (cationic dye)	FTIR,SEM,TGA	2012
27	PVC		Aliquate336	Cd(II),Cu(II),Zn(II)	SEM	2012
26	CTA	NPOE	1-decyl imidazole	Ni(II),Co(II),Cd(II),Zn(II)	-	2012
57	PVC		Aliquate336	Various salt of hydrochloric acid	-	2012
	CTA	NPPE	Alkyl derivative of resorcinarene	Pb(II),Zn(II),Cd(II)	-	2012
36	SLM & PIM		1-hexyl-2-Methylimidazole	Cu(II),Zn(II),Co(II),Ni(II)	AFM	2012
48	CTA	NPOE,TBF,TEHP,DOP	Calix [4]-Bis-2,3-Neptho-crown6(CNC)	Cs(I)	TIMM,AFM	2012
50	PVC	NPOE	D2EHPA	Ur(VI)	-	2012
51	CTA	NPPE	Cynex471X	Ag(I),Cu(II)	-	2012
35	CTA	NPOE	D2EHPA,TOPO	Pb(II),Cd(II)	FTIR,XRD,SEM	2011
33	CTA	NPPE	1-alkylimidazole	Cu(II),Zn(II),Co(II),Ni(II)	AFM	2011
53	PIM		5-(4-phenoxyphenyl)-6H-1,3,4-thiadiazine-2-amine	Cr(VI),Cu(II),Ni(II),Al(III), Fe(III)	AFM,FTIR	2011
54	PIM	Ionic liquid plasticizers 1-alkyl-3-methylimidazolium hexaflurophosphate or Tetrafluro Borate	CYPHOSIL104	Cr(VI)	-	2011

Table2

Reference No.	Membrane constituents	Carrier molecule	Target ions	Remarks/ year
13	Chloroform	Oxo crown ethers	Alkali and alkaline earth metals	2015
58	Di-chloromethane, chloroform, 1,2dichloroethane	Tripropyleneglycolmono butyl ether	Li(I), Na(I), K(I), picrates, dinitro phenolates, ortho nitro phenolates	2015
10	Chloroform	Tri butyl phosphate	Acetaminophen	2015
23	Fresh cooking oil, waste cooking oil, kerosene	D2EHPA & TBP as modifier	Cu(II)	2014
8	BLM	Calix [8]arene octa carboxylic acid	Methylene blue	2014
59	BLM	Schiff base (6,6-diethoxy-2, diylbis (nitrilomethylidene))-diphenol	Cu(II), Zn(II),Pb(II), Co(II), Ni(II), Ca(II), Mn(II), Fe(II), Hg(II)	2014
28	Chloroform	Diaza-18-crown-6	Alkai metals	2012
29	Chloroform	1,4,8,11-tetraazacyclotetradecane	Pb(II)	2012

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