Selective separation of radio-cesium from acidic solutions using supported liquid membrane containing chlorinated cobalt dicarbollide (CCD) in phenyltrifluoromethyl sulphone (PTMS)

P.K. Mohapatra, A. Bhattacharyya, V.K. Manchanda*

Radiochemistry Division, Bhabha Atomic Research Centre, Trombay, Mumbai 400085, India

ABSTRACT

A supported liquid membrane method was developed using chlorinated cobalt dicarbollide (CCD) in phenyltrifluoromethyl sulphone (PTMS) as the carrier, impregnated in PTFE flat sheet membranes for the selective separation of Cs(I) from nitric acid feed solution. Solvent extraction studies were carried out for optimizing the feed as well as strip conditions. >95% Facilitated transport of Cs(I) was observed in about 3 h when 1 M HNO_3 and 8 M HNO_3 were used as the feed and strip solutions, respectively while 0.025 M CCD in PTMS was used as the carrier extractant. Selectivity studies, carried out using a mixture of radiotracers viz. [51]Cr, ^{59}Fe, [99]Mo, ^{99m}Tc, [106]Ru, ^{137}Cs, [152]Eu and [241]Am, indicated selective transport of Cs(I) with DF values >100. Though reproducibility of the transport data was excellent when carried out in two successive transport experiments with freshly loaded carrier solvent, the stability of the membrane was poor which restricts its long term use.

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1. Introduction

The global yearly inventory of radioactive cesium produced in various nuclear reactors is approximately 16.8 tonnes [1,2]. Out of the three radioactive isotopes of Cs present in the high level waste ([134]Cs, [135]Cs and ^{137}Cs) ^{137}Cs is of great significance due to its reasonably long half life (30.1 y) and high yield. It has potential application as an excellent alternative source in gamma irradiators which are in use for the environmental pollution control, food preservation and sterilization of medical accessories. Industrial scale utilization of ^{137}Cs as a radiation source in irradiators requires that an effective separation process is available for the recovery of this fission product in mega curie quantities from the high level waste. Composition of a typical PHWR- high level waste is given in Table 1. Removal of ^{137}Cs from nuclear waste facilities is also helpful for vitrification and subsequent safe disposal of the high level waste (HLW) in deep geological repositories [1]. Amongst the separation techniques employed for Cs removal from waste solutions, solvent extraction has been particularly attractive because of its rapidity, ease of operation and large throughput [2]. Though there are solvent extraction methods involving exotic reagents such as the crown ethers, calixarenes and calix crowns for selective Cs recovery from the acidic waste solutions, chlorinated cobalt dicar-

bollide has been found to be particularly promising for the large scale processing of high level waste [3–5].

Though the first use of cobalt dicarbollide as an extractant for Cs was suggested by Rias et al. in the late 1970s [6], its use for the recovery of radio cesium from acidic wastes using the chlorinated derivatives in nitrobenzene was reported in the early nineties [7,8]. Nitrobenzene as the diluent has serious safety related issues which prompted search for alternative diluents. Of late, interest in the Cs recovery from HLW using chlorinated cobalt dicarbollide (CCD) has been renewed with the advent of UNEX (Universal Solvent Extraction process) and related processes [9–11]. Numerous laboratory batch studies with simulated and actual waste solutions, as well as counter-current pilot-scale flow sheet tests with simulated waste have been performed in INEEL (Idaho National Engineering and Environmental Laboratory), USA.

In recent years, membrane based methods have drawn the attention of separation chemists not only due to the low inventory of extractant and low energy requirement, but also due to the possibility of simultaneous extraction and stripping. Unfortunately, there is very little information available in the literature on Cs separation from acidic wastes based on liquid membrane separation methods [12–14]. We have used crown ethers as the Cs(I) selective extractant in our supported liquid membrane [13] and polymer inclusion membrane [14] studies for selective transport of the metal ion. However, the transport efficiency was not encouraging though excellent selectivity was observed. On the other hand, the available literature report lacked a systematic

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* Corresponding author. Fax: +91 22 25505151.
  E-mail address: vkm@barc.gov.in (V.K. Manchanda).

Table 1
Composition of high level waste (HLW) for pressurized heavy water reactor (PHWR); Acidity: 3 M HNO_3.

Constituent	Concentration (mg.l^{-1})	Constitutent	Concentration (mg.l^{-1})
Sr	186.3	Sb	4.7
Rb	74.5	Se	12.3
Zr	771.3	Dy	2.0
Ag	18.6	Sn	15.6
Ba	308.8	Te	102.8
Cd	16.3	Sm	163.8
Ce	532.5	Tb	5.0
Cs	543.8	Gd	165
Fe	500	Eu	22.6
Cr	100	Pr	243.8
Rh	127.5	Nd	862.5
Na	3000	La	263.8
Ni	100	Pd	267.5
U	18325	Ru	463.8
Tc	181.3	Y	99
Mo	731.3		

approach and, more work was needed especially in the area of membrane stability which was required for large scale continuous operations. An attempt has been made in the present work to develop a membrane based method for the selective separation of Cs-137 from solutions of acidic origin (such as the HLW) using CCD in phenyl trifluoromethyl sulphone (PTMS) as the carrier solvent.

2. Experimental

2.1. Materials

Chlorinated cobalt dicarbollide (CCD) was procured from Katchem, Czech Republic and was used without further purification. Radiotracers such as ^{51}Cr, ^{59}Fe, ^{99}Mo, ^{99m}Tc, ^{106}Ru, ^{137}Cs, ^{152}Eu were procured from Board of Radiation and Isotope Technology (BRIT), Mumbai and their radiochemical purity was ascertained by gamma spectrometry employing a high resolution HPGe detector. ^{241}Am was purified from aged plutonium samples following a reported method and its purity was determined by alpha-/gamma-spectrometry [15]. Phenyl trifluoromethylsulphone (PTMS) was synthesized from thioanisole through phenyltrifluoromethyl sulphide which was subsequently oxidized following a literature report [16]. The purity of PTMS was found to be >97%. All the other reagents used were of AR grade.

2.1.1. Preparation of CCD solution
CCD solution was prepared by dissolving the required quantity of the reagent in PTMS by sonicating in an ultrasonic bath. As the commercial CCD was available in the Cs$^+$-form, it was required to convert it to the H$^+$-form by equilibrating with 1:1 HClO$_4$ by about six times. Attempts to convert CCD to the H$^+$-form by equilibrating with HNO$_3$ or HNO$_3$ in *iso*-propanol was not successful as only partial conversion was possible as indicated by low D_{Cs} values.

2.1.2. PTFE membrane support
Microporous polytetrafluoroethylene (PTFE) membranes (diameter: 47 mm; pore size: 0.45 μm; thickness: 85 μm and porosity: 72%) obtained from Sartorius, Germany, were employed as the solid support for liquid membrane studies. The effective membrane area (Q), computed by multiplying the geometrical area and membrane porosity, was equal to 3.14 cm^2. Thickness and porosity of the membrane were ascertained as reported earlier [17].

2.2. Solvent Extraction Studies

Liquid-liquid extraction studies involving ^{137}Cs radiotracer were carried out in a manner reported by us earlier [18]. 0.5 mL organic solutions containing desired concentration of CCD (usually 0.025 M) in PTMS were agitated (by clamping on to rotating plates operated at a speed of 30 rpm) along with equal volumes of aqueous phases at the acidity of interest (containing the requisite quantity of ^{137}Cs tracer) in a thermostated water bath for an hour at 25.0 ± 0.1 °C. The two phases were then centrifuged and assayed by taking suitable aliquots from both the phases. Assay of ^{137}Cs was carried out by gamma counting using a well type NaI(Tl) scintillation detector inter phased to a multi channel analyzer. The distribution ratio (D_{Cs}) was calculated as the ratio of the concentration of the metal ion (expressed in terms of counts per unit time per unit volume) in organic phase to that in the aqueous phase. The material balance was within the experimental error limit of ±5%.

2.3. Transport Studies

The flat sheet supported liquid membrane (FSSLM) transport studies were carried out by using a specially fabricated Pyrex glass cell consisting of two compartments of 20 mL capacity connected by glass flanges which embedded the PTFE membrane containing the carrier solution. The microporous PTFE membranes were soaked in the carrier solution long enough to ensure that the membrane pores were filled with the carrier solution. Subsequently, the submerged membrane was removed from the solution and wiped carefully with a tissue paper to clear it of the excess carrier solution adhering to the surface of the support. The two phases were stirred at 400 rpm (for minimizing the aqueous diffusion layer thickness) using high speed magnetic stirrer equipped with precise speed control. Assay of ^{137}Cs in the feed as well as in the receiver phase was carried out at different time intervals to calculate the flux as well as permeability coefficient (*vide infra*). The co-transport of nitric acid from the feed to the strip compartment was also monitored by volumetric titrations using standard sodium hydroxide solution with phenolphthalein indicator. All the transport studies were carried out at ambient temperature (24 ± 1 °C). The material balance in these studies was found to be within the relative standard deviation of ±5%.

2.4. Transport Equations

Supported liquid membrane studies have indicated that the facilitated transport process is usually governed by a combination of three diffusion processes viz., diffusion across the aqueous diffusion layer in the feed phase, diffusion across the membrane phase and diffusion across the aqueous diffusion layer in the receiver phase (Fig. 1). The nature and the concentration of the carrier and the feed phase composition usually decide the distribution coeffi-
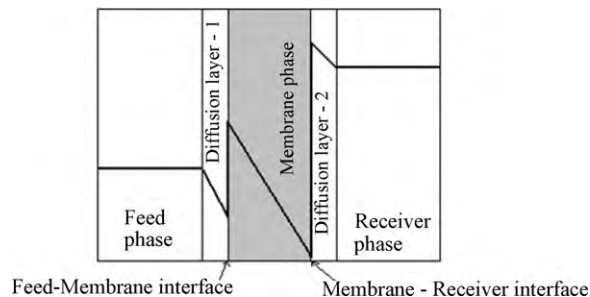
Fig. 1. A schematic representation of the transport mechanism indicating different diffusion layers and metal concentration profiles.

cient of the metal ion at the feed–membrane interface. Similarly, distribution coefficient values at the membrane–receiver phase interface decide the stripping efficiency. Though the extraction and the stripping distribution ratios and the kinetics of these processes are also important, the rate determining factor is usually the diffusion across the thickness of the membrane. The flux, across the membrane phase, is given by the Fick's first law of diffusion:

$$J_o = \frac{D_o([Cs^+.CCD^-]_{f,o} - [Cs^+.CCD^-]_{o,s})}{d_o} \tag{1}$$

where D_o is the membrane diffusion coefficient of Cs-CCD complex ($Cs^+.CCD^-$), d_o the membrane thickness, the concentration of Cs complex at the aqueous feed-membrane and membrane-aqueous strip interfaces are denoted by $[Cs^+.CCD^-]_{f,o}$ and $[Cs^+.CCD^-]_{o,s}$, respectively. The overall flux is related to the permeability coefficient, P, and the concentration of Cs in the feed as:

$$J = P.[Cs]_f \tag{2}$$

Where $[Cs]_f$ is the concentration of the metal ion in the feed. The flux can alternatively be expressed as

$$J = -(1/Q).dV_f[Cs]_f/dt \tag{3}$$

Where Q is the effective transport area (Q is equal to $A\varepsilon$ where A is the geometrical surface area and ε is the porosity) and V_f is the average volume of the feed compartment. Combining these two equations and integrating one obtains,

$$\ln([Cs]_{f,t}/[Cs]_{f,0}) = -(Q/V_f)P_f t \tag{4}$$

Where $C_{f,0}$ and $C_{f,t}$ represent the concentration of feed at starting time and after time 't', respectively. A plot of $\ln([Cs]_{f,t}/[Cs]_{f,0})$ *versus* time allows to evaluate the permeability coefficient from the slope of the linear fit. It should be noted that the above equation is valid only when carrier is not saturated and the flux linearly decreases with time. In the present work, since all the experiments were carried out at tracer metal concentration, Eq. (4) was applied for the calculation of permeability coefficient. The cumulative percent transport (%T) at a given time is represented by the equation:

$$\%T = 100(C_{f,o} - C_{f,t})/C_{f,o} \tag{5}$$

3. Results and Discussion

3.1. Distribution studies

Solvent extraction studies were carried out to optimize the experimental conditions for the subsequent supported liquid membrane studies. Herbst et al., have reported reasonably high extraction coefficients with 0.08 M CCD in fluorinated solvents [19]. As our high level waste solutions contained two different concentrations of nitric acid, 1 M HNO_3 for the Trombay waste and 3 M HNO_3 for the Tarapur waste, it was required to understand the effect of nitric acid concentration on Cs extraction. As higher extraction may lead to inefficient stripping, low ligand (CCD) concentration was used for these studies. Typically, 0.025 M CCD was chosen as the extractant concentration for 1 M HNO_3 while 0.063 M CCD was chosen when the aqueous phase acidity was 3 M. Increasing the CCD concentration further could lead to inefficient stripping of the extracted Cs(I). The extraction data as depicted in Fig. 2 indicated a steep decline in the D_{Cs} values with increasing nitric acid concentrations. Similar extraction behaviour was reported by Herbst et al. [19]. The effect of CCD concentration on Cs extraction was also investigated at 1 M HNO_3. A linear dependence of D_{Cs} on CCD concentration (Fig. 3) was observed with a slope of 1.00 ± 0.02 suggesting that the extracted ion-pair species is $(Cs^+ \cdot CCD^-)_{(org)}$ which is stabilized in a polar medium such as PTMS which has
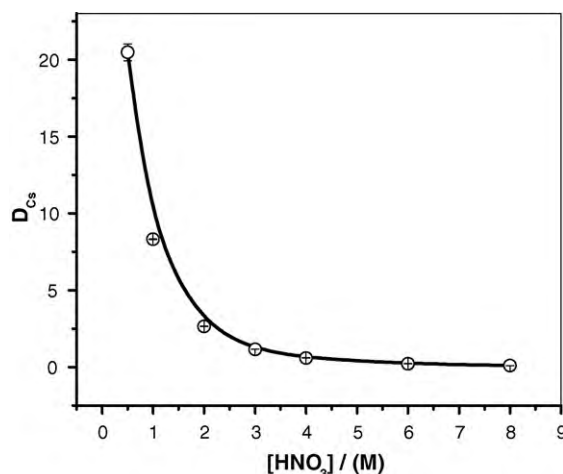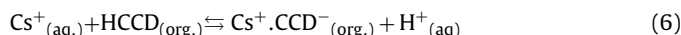
Fig. 2. Dependence of Cs extraction on aqueous phase nitric acid concentration. Organic phase: 0.025 M CCD in PTMS. Equilibration time: 60 minutes; Temp. = $25 \pm 0.1\,°C$.

reasonably high dielectric constant. The two-phase extraction equilibrium is represented as:

$$Cs^+_{(aq.)} + HCCD_{(org.)} \leftrightarrows Cs^+.CCD^-_{(org.)} + H^+_{(aq)} \tag{6}$$

where the subscripts (aq.) and (org.) represent the aqueous and the organic phases, respectively.

3.2. Liquid Membrane Studies

The flat sheet supported liquid transport data for Cs(I) permeation across the liquid membrane phase for a feed containing 1 M or 3 M HNO_3 and 0.025 M or 0.063 M CCD in PTMS as the carrier extractant are presented in Fig. 4. It takes about 90 minutes for >90% transport which is distinctly larger than to the transport rate reported by us from the SLM transport data using a Cs selective calix-crown ligand [20,21]. The plots showing the concentration profiles in the form of $[Cs]_t/[Cs]_0$ from feed as well as the receiver
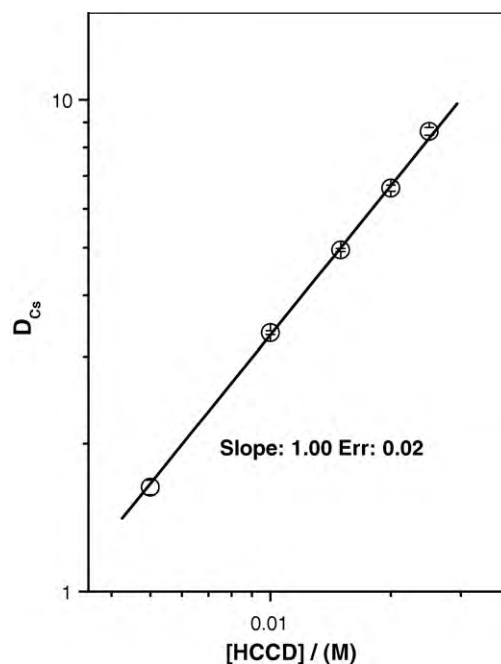
Fig. 3. Dependence of D_{Cs} on CCD concentration. Aqueous phase: 1 M HNO_3. Equilibration time: 60 minutes; Temp. = $25 \pm 0.1\,°C$.
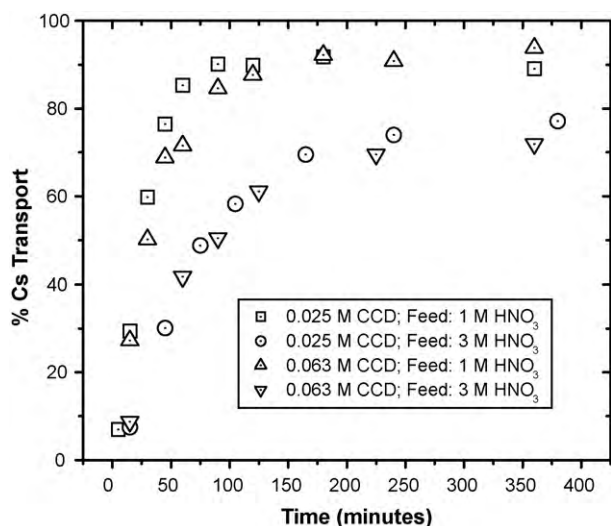
Fig. 4. Transport of Cs(I) by 0.45 μm PTFE flat sheet supported liquid membrane at varying feed and carrier concentrations. Strip: 8 M HNO₃ .

phases are shown in Fig. 5. The reproducibility of transport profile was checked by carrying out the transport studies under identical conditions using freshly loaded carrier solvents. The plots of the concentration profiles for two identical experiments are superimposed in Fig. 5. Excellent agreement suggests the reproducibility of the transport experiments.

3.2.1. Effect of carrier/nitric acid concentration on Cs(I) transport

As mentioned in Section 2.4 above, the transport of the metal ion is facilitated by the formation of organophilic species by the carrier which can diffuse across the membrane thickness. In the present case, the bulky CCD anion with high molar volume and low charge to volume ratio makes the extracted ion pair species highly organophilic. The transport model also indicates that higher transport rate is possible by increasing the extractant concentration. As indicated in eq. (6), an increase in the organic phase concentration of CCD is expected to increase (Cs⁺.CCD⁻) concentration at the membrane–feed interface. Similarly, increasing the feed phase acidity is expected to decrease the Cs transport rates. The effect of carrier concentration at two different feed nitric acid concentration (1 M & 3 M HNO₃) on Cs(I) transport is shown at two different CCD concentrations in Fig. 4. Contrary to our expectation, the transport rates at 0.063 M CCD concentration were lower than those
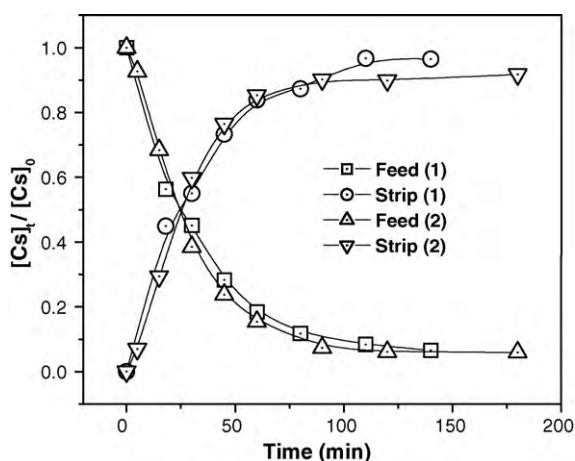

Fig. 5. Transport of Cs(I) through 0.45 μm PTFE flat sheet supported liquid membrane; Carrier: 0.025 M CCD in PTMS; Feed: 1 M HNO₃ ; Strip: 8 M HNO₃.

Table 2
Transport parameters for the Cs(I) permeation across supported liquid membrane containing chlorinated cobalt dicarbollide in PTMS as the carrier extractant and 8 M HNO₃.

Feed	Carrier extractant	% Transport (90 min.)	$P \times 10^3$ (cm.s^{-1})
1 M HNO₃	0.025 M CCD in PTMS	90.1	2.95 ± 0.29
3 M HNO₃	0.025 M CCD in PTMS	54.3	0.79 ± 0.07
1 M HNO₃	0.063 M CCD in PTMS	84.6	1.79 ± 0.19
3 M HNO₃	0.063 M CCD in PTMS	50.5	0.81 ± 0.07

Table 3
Density and viscosity data of the carrier solutions.

Carrier solution	Density (g.cm^{-3})	Viscosity (m.Pas^{-1})
0.025 M CCD in PTMS	1.3972	3.7477
0.063 M CCD in PTMS	1.4003	4.1750

observed for 0.025 M CCD. This is clearly seen in the permeability coefficient values, calculated using eqn. (4), which are listed in Table 2. Also, the transport rates, as expected, were significantly lower when the feed acidity was increased from 1 M HNO₃ to 3 M HNO₃. The permeability coefficient value for 0.025 M CCD in PTMS as the carrier, decreased 3–4 times when the feed acidity was increased from 1 M to 3 M HNO₃ (Table 2). Interestingly, the P value decrease was <50% of the permeability coefficient for similar increase in the feed acidity when the carrier concentration was 0.063 M CCD in PTMS. This could be attributed to either inefficient stripping at the membrane–receiver interface or due to slower diffusion of the extracted species. As the extracted species remained same over the CCD concentration range (Fig. 3), the diffusion is directly related to the viscosity of the carrier solution (on the basis of the Stokes–Einstein equation) on increasing from 0.025 M to 0.063 M. Viscosity and density data (Table 3) of the carrier solutions indicated no significant difference between the two concentrations of the carrier solutions employed. On the other hand, the D$_{Cs}$ value at the membrane–receiver interface is determined to be 0.31 (at 8 M HNO₃) for 0.063 M CCD as compared to a D$_{Cs}$ value of 0.1 (at 8 M HNO₃) for 0.025 M CCD suggesting poor stripping is the reason for the inferior transport behaviour when 0.063 M CCD is used as the carrier (Table 4). This indicated that the effect of distribution coefficient plays a significant role as compared to the diffusion coefficient. These studies indicate that liquid membrane using CCD in PTMS is efficient for Cs recovery at ∼ 1 M HNO₃ as the feed acidity. This is relevant for HLW if it is stored at ∼ 1 M HNO₃.

The present work has been carried out using tracer concentration of Cs. The transport rate is likely to decrease with increase in Cs concentration. As indicated in Table 1, the concentration of Cs is the high level waste is quite significant and the flat sheet supported liquid membrane may take much longer time for quantitative transport. For such feed solutions, more efficient set up such as the hollow fiber supported liquid membrane may be used.

3.2.2. Selectivity studies using a mixture of radiotracers

Though transport of Cs was possible using 1 M HNO₃ as the feed and 8 M HNO₃ as the receiver solutions, it was required to investigate the selectivity of Cs transport using CCD as the car-

Table 4
Distribution coefficient data for Cs(I) at different feed acidities at different concentrations of CCD.

[HNO₃], M	D$_{Cs}$ at different CCD concentrations [CCD] = 0.025 M	[CCD] = 0.063 M
1	8.32 ± 0.01	23.6 ± 0.2
3	1.15 ± 0.02	3.77 ± 0.06
8	0.10 ± 0.01	0.31 ± 0.02
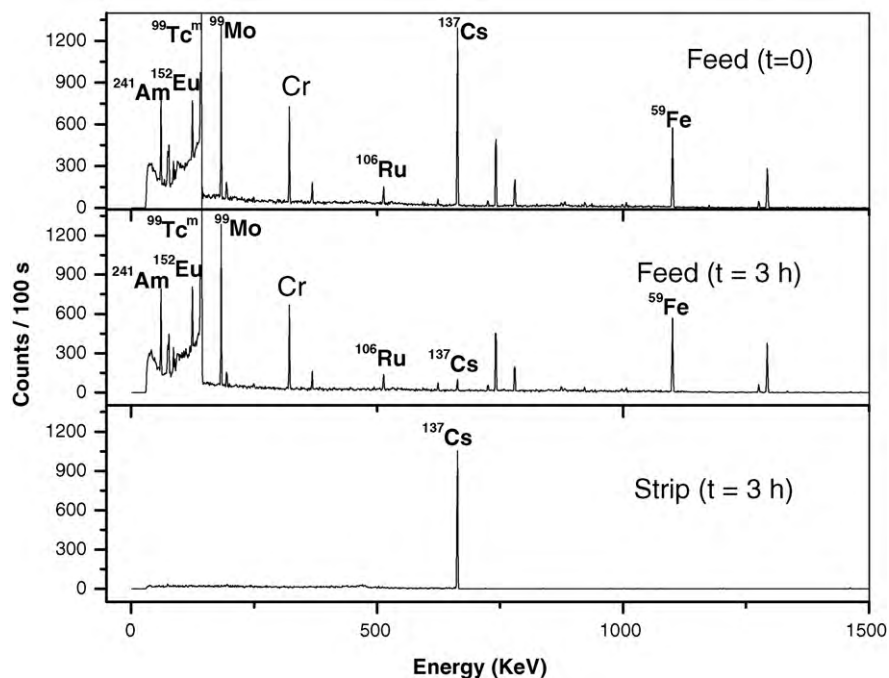
Fig. 6. Gamma spectrum of the radionuclides in the feed, strip and raffinate solutions showing the selective transport of ^{137}Cs through 0.45 μm PTFE flat sheet supported liquid membrane after 3 h of operation; Carrier: 0.025 M CCD in PTMS; Feed: 1 M HNO$_3$; Strip: 8 M HNO$_3$.

rier extractant. This was done by preparing a synthetic mixture of radionuclides such as [99]Mo, ^{99m}Tc, [106]Ru, ^{137}Cs, [152]Eu (some representative fission products), ^{51}Cr, [59]Fe (some typical structural materials) and [241]Am in 1 M HNO$_3$ and using it as the feed solution. The transport data after 3 h of operation is presented in Fig. 6. Fig. 6 also includes the gamma ray spectrometry data of the feed data before the start of the experiment as well as after 3 hours of operation along with the strip phase data after 3 h. As indicated in the figure, excellent selectivity was observed with respect to the fission products, activation product and structural materials suggesting feasibility of the present method for selective recovery of Cs from HLW at 1 M HNO$_3$. The decontamination factor (D.F., defined as the ratio of product to impurity in the product divided by that in the feed) values are listed in Table 5. Though ^{99m}Tc lines are indicated in the figure, the D.F. values with respect to this radioisotope was not included as decay product of [99]Mo also contributed to the total ^{99m}Tc quantity. As indicated, the D.F. values are >100 for all the radionuclides studied in the present work suggesting this separation method is similar to the liquid membrane based separation methods reported earlier by us using calix[4]-bis-2,3-naphtho-crown-6 [20].

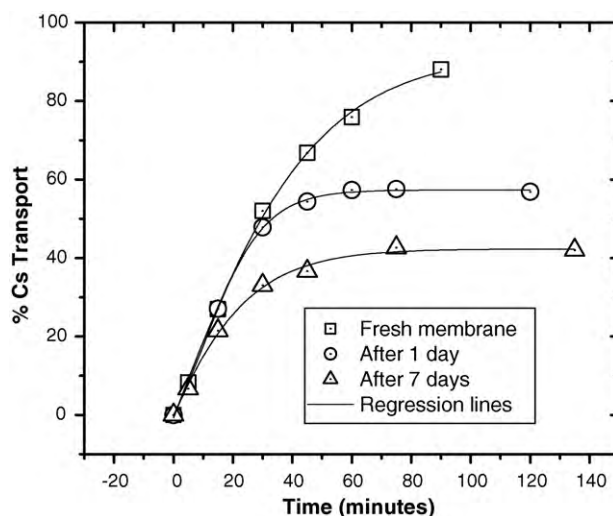### 3.2.3. Stability of the liquid membrane

The stability of flat sheet supported liquid membrane using CCD in PTMS as the carrier extractant was studied by using the same SLM in several runs over a period of 7 days. Fig. 7 shows the transport profiles of Cs(I) using the same SLM, after one day and after seven days. As shown in the figure, the transport rate decreased signifi-

cantly with time. The transport equation as given by eqn (4) shows a linear dependence of $\ln([Cs]_{f,t}/[Cs]_{f,0})$ on the transport time. However, with detorioration of the liquid membrane with time, deviation from linearity is expected. As shown in Fig. 8, such deviation is seen even after 1 day (where deviation from straight line behaviour was seen after 30 minutes) and is much more pronounced after 7 days of operation (deviation was clearly seen even at 15 minutes). The probable reasons responsible for the degradation of SLM is already reported in the literature by different research groups [22,23]. One of the reasons for the poorer performance of SLM is the loss of LM phase (carrier/solvent) from the pores of solid support. As indicated by Kemperman et al., the stability of the membranes is greatly influenced by the viscosity as well as miscibility of the carrier phase [23]. The diluent is also reported to play a major role in deciding

Table 5
Decontamination factors (D.F.) of ^{137}Cs over fission products, activation products and structural materials. Carrier: 0.025 M CCD in PTMS; Feed: 1 M HNO$_3$ containing the radiotracer mixture; Receiver: 8 M HNO$_3$.

Radio tracer	D.F.	Radiotracer	D.F.
[51]Cr	389 ± 15	[106]Ru	114 ± 15
[59]Fe	312 ± 12	[152]Eu	301 ± 21
[99]Mo	329 ± 13	[241]Am	232 ± 18

Fig. 7. Transport of Cs(I) through 0.45 μm PTFE flat sheet supported liquid membrane as a function of number of days of operation; Carrier: 0.025 M CCD in PTMS; Feed: 1 M HNO$_3$; Strip: 8 M HNO$_3$.

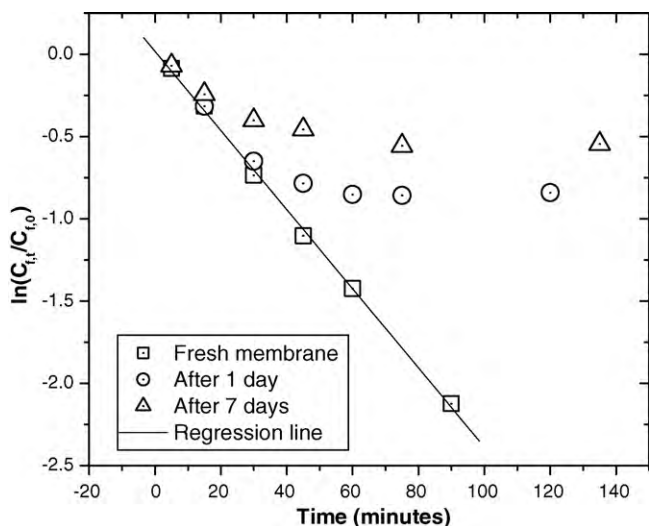**Fig. 8.** Comparison of SLM stability as indicated by the semi log plots of $\ln[C_{f,0}]/[C_{f,t}]$ vs time. Carrier: 0.025 M CCD in PTMS; Feed: 1 M HNO$_3$; Strip: 8 M HNO$_3$.

the membrane stability [24,13,25]. Coloration of the feed and the receiver phase after prolonged use suggested leaching out of the extractants from the membrane pores. Acid transport (counter-transport from receiver phase to the feed phase) data, as given in Fig. 9, shows increase in acid transport with number of days the membrane has been used. As this can not be explained on the basis of the facilitated transport theory which is valid for Cs transport, it can be ascribed to hydrophilic channel formation for acid counter-transport.

The SLM stability can be improved by opting for strip dispersion technique which can replenish the leached out solvent and carrier molecules from the pores [26]. Modifying the diluent can also improve the SLM stability. Alternatively, polymer inclusion membranes can be prepared wherein the loss of carrier molecules is minimized [27]. However, the flux can be significantly lower in case of the later membrane transport system.
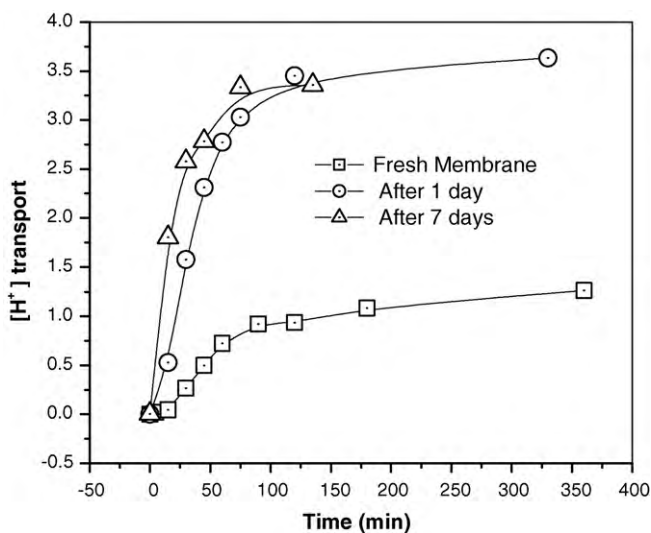
4. Conclusions

The transport behaviour of Cs(I) was investigated from nitric acid medium using chlorinated cobalt dicabollide (CCD) in phenyl trifluoromethyl sulphone (PTMS) as the carrier in microporous PTFE membranes. The transport rates were influenced by the feed acidity and an increase in the feed acidity resulted in a sharp decrease in the permeability coefficient value. Though increasing the carrier concentration is expected to result in higher transport rates an opposite trend was observed which was ascribed to poor stripping at the membrane–receiver interface. The selectivity studies have shown promise while the stability of the liquid membrane is of concern suggesting each time a fresh liquid membrane has to be used. Alternatively, diluent modification may be tried which may arrest the leaching of the extractants from the membrane pores. Strip dispersion technique can also be used for longer stability and hence reusability of this liquid membrane [26]. Polymer inclusion membrane may also be evaluated for Cs recovery from acidic wastes. The problem of stability needs to be addressed to enable its application for large scale recovery of Cs from HLW solutions maintained at 1 M HNO$_3$.

ABBREVIATIONS

CCD	Chlorinated cobalt dicarbollide
D.F.	Decontamination factor
HLW	High Level Waste
INEEL	Idaho National Engineering and Environmental Laboratory
PHWR	Pressurized Heavy Water Reactor
PTFE	Poly Tetra Fluoro Ethylene
PTMS	Phenyl trifluoromethyl sulphone
SLM	Supported Liquid Membrane
UNEX	Universal Extraction

SYMBOLS

J	Overall flux
J$_o$	Membrane flux
D$_o$	Membrane diffusion coefficient
d$_o$	Membrane thickness
[Cs]$_t$	Concentration of Cs in aqueous feed solution at time t
[Cs]$_0$	Initial concentration of Cs in aqueous feed
P	Permeability coefficient
Q	Effective surface area of membrane
A	Geometrical surface area of membrane
V$_f$	Time average aqueous feed volume

Fig. 9. Membrane stability data as indicated by the acid transport from receiver phase to the feed phase as a function of operation time. Carrier: 0.025 M CCD in PTMS; Feed: 1 M HNO$_3$; Strip: 8 M HNO$_3$.

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