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Abstract: Solvent extraction studies of Y^{3+} and Sr^{2+} with 2-ethylhexyl 2-ethylhexyl phosphonic acid (KSM-17) and octyl(phenyl)-N,N-diisobutylcarbamoylmethyl-phosphine oxide (CMPO) are carried out from aqueous media containing a wide range of nitric acid and other potential reagents to arrive at the operating conditions for the selective transport of ^{90}Y using supported liquid membrane (SLM) containing these reagents as carriers. Since the transport data of ^{90}Y using single cell SLM with KSM-17 was available from our earlier experiments, single cell transport studies with CMPO carrier are only carried out to optimize the strippant phase. Transport studies with pure ^{90}Y is carried out using a transport cell with two SLMs one with KSM-17 and the other CMPO carriers to optimize the transport parameters. Based on these data the development of a two stage SLM system for the generation of carrier free ^{90}Y from ^{90}Sr source is described. The procedure described is amenable for automation and scale up.

Keywords: ^{90}Sr-^{90}Y generator, supported liquid membrane, solvent extraction, PUREX HLLW, CMPO, KSM-17

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INTRODUCTION

Yttrium-90 is one of the important radionuclides for therapeutic use in nuclear medicine. Suitability of this isotope is because of its short half-life ($T_{1/2} = 64.2$ h) and high-energy beta emissions ($E_{max} = 2.28$ MeV). Its chemical properties are also suitable for forming stable bindings with various chelating agents (1–4). ^{90}Y can be prepared by neutron-irradiation of yttrium metal or its oxide. But this method does not produce carrier-free ^{90}Y, which is a serious drawback in therapeutic applications. Another source of ^{90}Y is through the decay of ^{90}Sr which is abundantly present in high level liquid waste (HLLW) solutions of PUREX origin. ^{90}Sr ($T_{1/2} = 28$ y) attains secular equilibrium with ^{90}Y in a short period and can be used as a perpetual source of ^{90}Y.

Methods based on ion-exchange, extraction chromatography, and solvent extraction either alone or in combination with each other have been reported in the literature for the separation of carrier-free ^{90}Y for radiotherapeutic application (5–11). The most important concern is the radiochemical purity of the ^{90}Y product especially from ^{90}Sr as it is a bone seeker. Earlier reports (9–11) from this laboratory had described the procedure for the separation ^{90}Sr in mCi levels from HLLW by suitably modifying the conventional radiochemical method (11) and the use of the recovered pure ^{90}Sr for the separation of ^{90}Y using single stage supported liquid membrane (SLM) technique employing 2-ethylhexyl 2-ethylhexyl phosphonic acid (KSM-17) as carrier. The product was separated as YCl_3 or $Y(NO_3)_3$ in 1 M HCl/HNO_3 medium depending on the requirement. The transport period of about 3–4 hours was used for ^{90}Y separation. Under recommended conditions, the yield of separation was found to be only $\sim$40%, which could be enhanced by changing the acidity of the feed/receiver phase appropriately or by increasing the transport period. Invariably, higher yield of ^{90}Y also enhances the ^{90}Sr contamination in the product making it unsuitable for radiotherapeutic applications.

Therefore further studies are carried out to explore the possibility of getting pure ^{90}Y in higher yield. Additionally, an attempt to obtain ^{90}Y in acetate form is also made, since acetate medium is preferred over nitrate or chloride for radiolabeling of bio-molecules. Two-stage SLM based separation of ^{90}Y from ^{90}Sr reported in this paper is principally based on their solvent extraction properties of two organophosphorous extractants *viz.* 2-ethylhexyl 2-ethylhexyl phosphonic acid (KSM-17-acidic) and octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide (CMPO-neutral) under optimum conditions. Extraction data with strontium and yttrium from conventional solvent extraction studies using these solvents under different experimental conditions are collected and used in designing the new separation system. Using the extraction data of the two solvents as a guide, further studies have been carried out to optimize the transport of ^{90}Y and ^{90}Sr using single stage as well as two stage transport cells to select the best transport conditions for ^{90}Y without permeation of ^{90}Sr in the final product. Since transport data for

the single stage SLM cell containing KSM-17 as carrier is readily available (10, 11) more attention was focused on single stage SLM cell employing CMPO as carrier. The improvement in the radiochemical purity of ^{90}Y over the earlier method is brought about by incorporating a second stage membrane with carrier phase consisting of octyl(phenyl)-N,N-diisobutylcarbamoylmethyl phosphine oxide (CMPO). The two membranes are used in two different modes *viz.* simultaneous and sequential modes. Among the various receiving media studied, 1 M acetic acid is selected as the receiver phase for getting the product in acetate medium. The yield of separation is observed to be better than $95 \pm 3\%$.

EXPERIMENTAL

Materials

Commercially available polytetrafluoroethylene (PTFE) membranes are used in all the experiments. Membranes have an average pore diameter and thickness of 0.45 μm and 160 μm, respectively. The porosity of the membrane is about 84%. Carriers, 2-ethylhexyl 2-ethylhexyl phosphonic acid (KSM-17) and octyl(phenyl)-N,N-diisobutylcarbamoylmethyl phosphine oxide (CMPO) are synthesized and purified at this research centre using the procedures reported in the literature (12, 13). n-dodecane (~93% C-12) procured from M/s. Transware Chemia Handelsgeselschaft Hamburg, Germany is used as diluent. All other acids and reagents used are of analytical reagent grade.

Radioactive Tracers

The radioactive tracer $^{85+89}Sr$ is procured from the Board of Radiation and Isotope Technology, Department of Atomic Energy (India) and ^{90}Y is obtained from a single stage SLM generator system developed in-house (10). $^{90}Sr/^{90}Y$ mixture is obtained from the purified lot of ^{90}Sr recovered earlier (11).

Solvent Extraction Studies in Batch

Batch extractions experiments are carried out from different aqueous media such as nitrate, chloride and acetate in order to study the extraction behavior of strontium and yttrium independently using appropriate concentrations of the carriers. In these experiments 2 mL of the aqueous phase at the desired acid concentrations containing tracer quantity of the radionuclide is contacted with equal volume of the organic phase till equilibrium is attained. The phases are separated and assayed for their activity. Extraction data is expressed as distribution ratio. The extraction is carried out from a wide range of nitric acid concentration varying from molar to pH range for

both the solvents. These data are used in selecting the optimum conditions for extraction/stripping as well as in selecting a suitable receiver phase.

Impregnation of Carrier in PTFE Membrane

The PTFE membranes are impregnated with KSM-17 (undiluted) and different concentration of CMPO in n-dodecane separately by immersing them in a desired concentration of the reagents for at least 12 hours before use. The membranes are rinsed with water prior to the start of the transport experiments.

Supported Liquid Membrane Studies

Single stage permeation measurements across SLM are carried out with a two compartment permeation cell consisting of a feed chamber and a receiver chamber with a volume of 5.0 mL each, separated by a SLM having an effective membrane area of 1.13 cm^2. In the two stage membrane system in simultaneous mode, three compartment cells are used with two SLMs separating the feed and intermediate compartment (with KSM-17 supported PTFE membrane) whereas the other separates the intermediate and final product compartments (using the CMPO supported PTFE membrane). The solutions in each compartment are mechanically stirred using a magnetic stirrer to minimize the thickness of the aqueous diffusion layer. The feed solutions used during the transport studies contain either pure $^{85+89}Sr$ tracer or ^{90}Y in desired concentrations of nitric acid. The adjustment of pH is done by drop-wise addition of either acid or alkali (1 M HNO_3 or 1 M NaOH).

Analysis

Suitable aliquots (generally 20 μL) of the samples are drawn from each compartments at different intervals and are assayed for beta activity using a β- proportional counter standardised against a RaDE source. A NaI(Tl) scintillation counter standardised against ^{137}Cs source is used for gamma activity. Individual gamma emitting fission products are assayed with a 62 cc HPGe detector coupled to a 4 K multi-channel analyser.

RESULTS AND DISCUSSION

Solvent Extraction Studies of ^{90}Y and ^{90}Sr using KSM-17 and CMPO

Batch extraction experiments are carried out to study the behavior of Sr and Y from nitric acid medium using 20% KSM-17 in n-dodecane and 0.2 M CMPO

in n-dodecane as extractant. The extraction is studied from the nitric acid medium in the concentration range of 4.0 M to pH 3 for acidic extractant KSM-17 whereas acidity of the aqueous phase is varied from 4.0 to 0.1 M HNO_3 in the case of neutral extractant CMPO.

Extraction data obtained using KSM-17 are given in Table 1. In general, the extraction of yttrium is much higher compared to that of strontium and increased substantially with decrease in acidity. Practically negligible extraction of Sr^{2+} is observed from feed solutions with nitric acid concentration in the range 4 M to pH 1 ($D_{Sr} < 10^{-3}$ for all). At higher pH, the distribution ratios are of the order of 10^{-3}. As expected, Y extraction increased with decreasing concentration of HNO_3. Using the distribution ratios under identical conditions, separation factors are calculated and are listed in the same table. Results indicate that at lower concentration of HNO_3 (pH 2 and 3) the separation factors (D_Y/D_{Sr}) obtained are of the order of 10^4. Similar results using CMPO as extractant are given in Table 2. These results show that as the concentration of HNO_3 increases, D_Y also increases. At all the concentrations of HNO_3 the extraction of strontium is negligible.

These data suggest that KSM-17 does not extract strontium when present in acidic aqueous solutions in the concentration range of 4 M HNO_3 to pH 3. Extraction of yttrium by KSM-17 is nearly as poor as strontium under highly acidic conditions (0.5–4 M HNO_3) while it is high in the pH range 1–3 giving separation factors of the order of 10^4.

In the case of CMPO, under acidic conditions of 0.1-4 M HNO_3 strontium extraction is insignificant while yttrium extraction increases with increasing concentration of HNO_3. Y^{3+} when present in high concentration of nitrate gets extracted into the CMPO containing organic phase, which can be back extracted or stripped into aqueous phase with lower concentration of nitrate.

Table 1. Extraction of Sr^{2+} & Y^{3+} from HNO_3 medium using KSM-17

Experimental parameters
Feed activity: ^{90}Y (β) = 73290 Bq/mL, $^{85+89}Sr$ (γ) = 49712 Bq/mL,
Extractant: 20% KSM-17 in n-dodecane, Phase ratio: 1:1,
Volume: 2 mL, Contact time: 15min.

$[HNO_3]$ in feed	Extraction of ^{90}Y (D_Y)	Extraction of $^{85+89}Sr$ (D_{Sr})	Separation factor $(D_Y)/(D_{Sr})$
4.0 M	<0.01	$<10^{-3}$	—
2.0 M	0.04	$<10^{-3}$	>40
1.0 M	0.07	$<10^{-3}$	>70
0.5 M	0.35	$<10^{-3}$	$>3.50 \times 10^2$
pH 1	2.66	$<10^{-3}$	$>2.66 \times 10^3$
pH 2	116.57	2.70×10^{-3}	4.32×10^4
pH 3	126.40	4.40×10^{-3}	2.87×10^4

Table 2. Extraction of Sr^{2+} and Y^{3+} from HNO_3 medium using CMPO

Experimental parameters
Feed activity: ^{90}Y (β) = 29740 Bq/mL, $^{85+89}Sr$ (γ) = 31290 Bq/mL,
Extractant: 0.2 M CMPO in n-dodecane, Phase ratio: 1:1,
Volume: 2 mL, Contact time: 15min.

$[HNO_3]$ in feed	Extraction of ^{90}Y (D_Y)	Extraction of $^{85+89}Sr$ (D_{Sr})	Separation factor $(D_Y)/(D_{Sr})$
4.0 M	3.47	$<10^{-3}$	$>3.47 \times 10^3$
3.0 M	2.80	$<10^{-3}$	$>2.80 \times 10^3$
2.0 M	2.17	$<10^{-3}$	$>2.17 \times 10^3$
1.0 M	1.48	$<10^{-3}$	$>1.48 \times 10^3$
0.5 M	0.804	$<10^{-3}$	$>8.04 \times 10^2$
0.1 M	0.163	$<10^{-3}$	$>1.63 \times 10^2$

In the case of acidic extractant, KSM-17, extraction of trivalent metal ions is favored at a lower concentration of nitric acid, which is stripped at a higher concentration of nitric acid.

In order to select a receiver phase for CMPO based SLM system, a few extraction experiments are carried out from different aqueous medium viz. HNO_3, HCl, HFC (a mixture of 0.4 M hydrazine hydrate +0.4 M formic acid +0.1 M citric acid) and CH_3COOH. The results are given in Table 3. Low value of distribution ratio indicates its use as a good strippant (receiver phase) for a metal ion under those conditions. The complexing mixture HFC has been used in our earlier studies (14, 15) as a stripping agent for trivalent actinides from the CMPO phase during the partitioning of

Table 3. Extraction of Y^{3+} and Sr^{2+} from different aqueous medium using CMPO

Experimental parameters
Feed activity: ^{90}Y (β) = 44955 Bq/mL, $^{85+89}Sr$ (γ) = 35621 Bq/mL,
Extractant: 0.2 M CMPO in n-dodecane, Phase ratio: 1:1,
Volume: 2 mL, Contact time: 15min.

Aqueous phase	Extraction of ^{90}Y (D_Y)	Extraction of $^{85+89}Sr$ (D_{Sr})
4.0 M HNO_3[a]	165.31	$<10^{-3}$
4.0 M HNO_3	3.47	$<10^{-3}$
1.0 M HNO_3	1.42	$<10^{-3}$
1.0 M HCl	0.07	$<10^{-3}$
HFC[b]	0.03	$<10^{-3}$
1.0 M CH_3COOH	0.04	$<10^{-3}$

[a]0.8 M CMPO in n-dodecane
[b]0.4 M hydrazine hydrate + 0.4 M formic acid +0.1 M citric acid

actinides from HLLW. Since the behavior of Y^{3+} is similar to that of trivalent actinides, this reagent is used as a strippant for Y from the loaded CMPO phase also. These results indicate that acetic acid, HCl as well as HFC are suitable strippants for ^{90}Y from the CMPO loaded organic phase. Using the above data as a guide, further studies are carried out to optimize the selective transport conditions of ^{90}Y from ^{90}Sr.

Transport Studies

Transport through Single Stage Membrane

Batch equilibrium studies have established the preferential extraction of Y^{3+} over Sr^{2+} as seen from the high separation factors achieved under the optimized conditions. Hence SLM studies with ^{90}Y alone were carried out for single stage transport. In SLM based separation, one of the parameters on which the transport rate of the metal ion depends is the concentration of the carrier. Our earlier experience in the use of CMPO as carrier shows that 0.8M CMPO in n-dodecane as the optimum concentration for the transport of trivalent metal ions from 3–4 M HNO_3 (16). Similar extraction behavior is found for $^{90}Y^{3+}$ also from the nitric acid medium. The transport of $^{90}Y^{3+}$ present in 4 M HNO_3 through SLM containing CMPO (at 0.2 M and 0.8 M CMPO in n-dodecane) into a receiver compartment containing 1 M CH_3COOH are presented in Fig. 1.

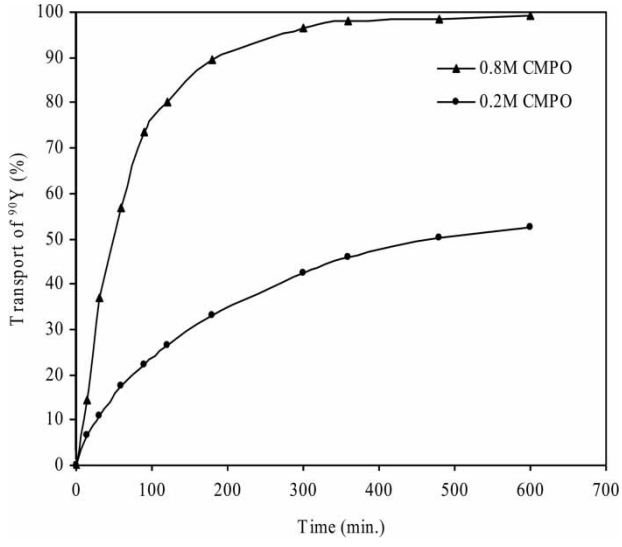

Figure 1. Transport of ^{90}Y through CMPO based SLM using acetic acid as receiver phase.

It is seen that as time increases, ^{90}Y transport increases. More than 90% of ^{90}Y can be transported within 4 hours when the carrier concentration is 0.8 M CMPO in n-dodecane as against ~40% with 0.2 M CMPO. Near quantitative transport is observed in about 5 hrs with 0.8 M CMPO. For higher rates of transport, 0.8 M CMPO in n-dodecane is used as carrier in further transport studies.

Transport of ^{90}Y from 4 M HNO_3 when different forms of acetate, such as acetic acid, sodium acetate, and ammonium acetate are used as strippants in the receiver compartment, separated by SLM with 0.8 M CMPO in n-dodecane, is studied with time and the results are presented in Fig. 2. Among the three, acetic acid is found to be the most efficient with >95% transport of ^{90}Y in about 5 hours. In the case of ammonium acetate, initially the transport rate is comparatively slow, but in about 7 hours the separation yield is almost equal to that of acetic acid. Sodium acetate shows about 90% transport under identical conditions. Based on these results, acetic acid is chosen as the strippant for further studies.

The effect of the concentrations of acetic acid in the receiving phase is also studied and it is seen that ~60% of ^{90}Y gets transported in ~6 hours when 0.5 M acetic acid is used while this increases to >95% with 1 M acetic acid within 5 hours (Fig. 3).

Transport through Two Stage Membrane

Transport of Pure ^{90}Y

Encouraged by the above results with single cell, transport studies using KSM-17 and CMPO as carriers in a two stage transport cell with three compartments

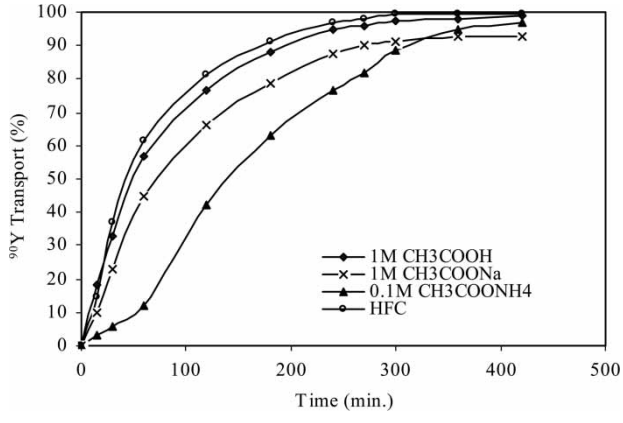

Figure 2. Transport of ^{90}Y from 4 M HNO_3 using different receiving phases.

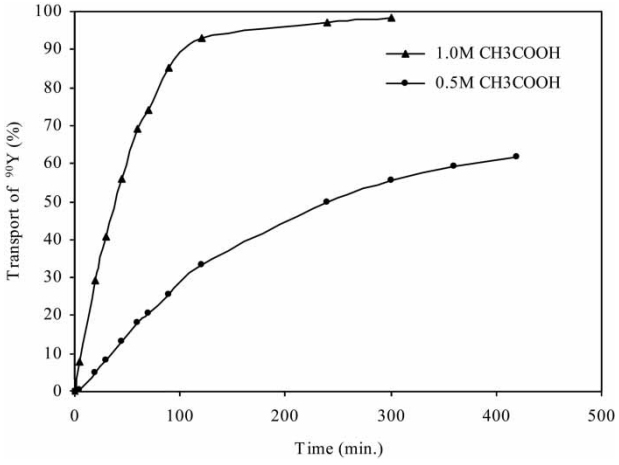

Figure 3. Effect of the [CH_3COOH] on the transport of ^{90}Y through CMPO based SLM.

of 5 mL capacity each are carried out. The schematic diagram of the cell is given in Fig. 4.

In these studies the transport of ^{90}Y is carried out using a two stage transport cell containing three compartments of capacity 5 mL each for feed, intermediate phase, and product respectively. Initially, pure ^{90}Y at concentration of ~420 MBq/L (11.36 mCi/L) is spiked in nitric acid (pH 2) and introduced in the feed compartment. 4 M nitric acid is used in the intermediate compartment and 1 M acetic acid in the product compartment. KSM-17 based SLM is used in between feed and the intermediate compartment whereas CMPO based SLM was used in between the intermediate and the final product compartment. The transport of ^{90}Y is monitored by following the β-activity in each compartment with time, which is depicted in Fig. 5.^{90}Y is

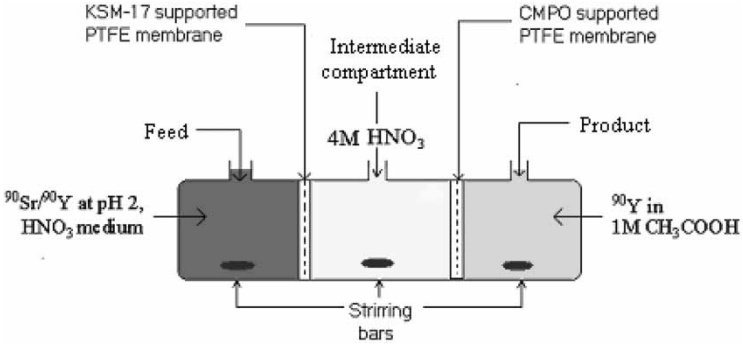

Figure 4. Schematic diagram of a two stage membrane cell.

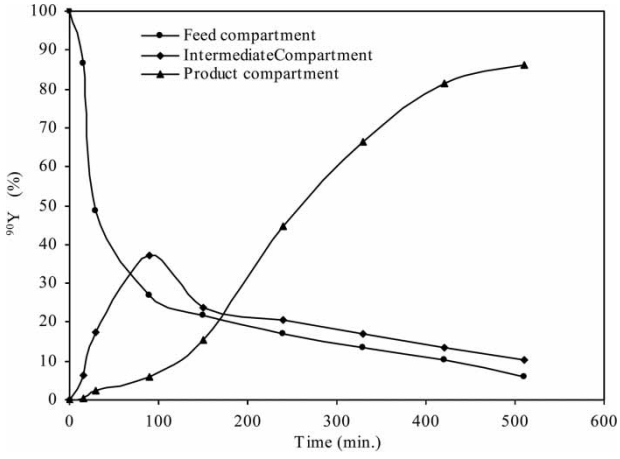

Figure 5. ^{90}Y profile in different compartments of a two stage cell.

found to be transported quickly in the beginning, as seen from the reduction of activity by >50%, within 30 minutes. As the concentration of the ^{90}Y in the feed compartment decreases, its rate of transport is also reduced. In about 7 hours >80% of the initial ^{90}Y activity is transferred to the product compartment.

Transport of ^{90}Y from ^{90}Sr/^{90}Y Mixture

Subsequently, the same two stage generator cell is used for the separation of carrier free ^{90}Y from ^{90}Sr-^{90}Y mixture. ^{90}Sr solution in equilibrium with ^{90}Y in HNO_3 adjusted to pH 2 is used in the feed compartment separated from the intermediate compartment containing 4 M HNO_3 by KSM-17 impregnated PTFE membrane. CMPO impregnated PTFE membrane separates the intermediate compartment from the third receiver compartment containing 1 M acetic acid. The passage of ^{90}Y monitored by β-activity in each compartment as a function of time and given in Fig. 6.

These results clearly indicate that quantitative transport of ^{90}Y from the feed compartment takes place within about 4 hours. Thereafter, the activity in the feed compartment remains constant. The β-activity in the intermediate compartment rises initially then decreases due to its transport through CMPO based SLM in the second stage to the product (receiver) compartment. However, the β-activity in the product compartment increases at a slower rate compared to the intermediate compartment and after about 7 hours, this rate decreases further. More than 40% of the total beta activity is transported in the product compartment in about 10 hours, which indicates >80% yield of ^{90}Y separation. This slow rate of

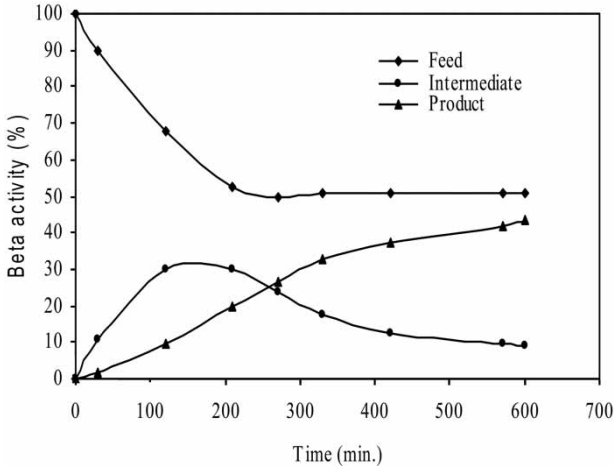

Figure. 6. Beta activity profile of each compartment of a two stage cell using ^{90}Sr-^{90}Y mixture as feed.

transport towards the end can be attributed to the changes in the concentration gradient.

For selective and faster transport of ^{90}Y, an alternative approach has also been worked out in which two single stage membrane cells are used sequentially (Fig. 7). ^{90}Y is efficiently transported through KSM-17 based SLM in the first stage. The product ^{90}Y in the second stage is further transported through CMPO based SLM. The SLM system operated in this mode has shown >95%

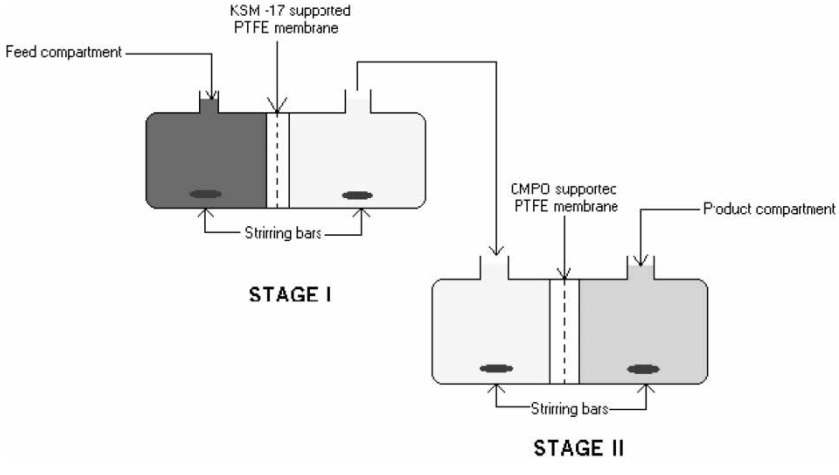

Figure 7. Schematic diagram of a two stage membrane cell in sequential mode.

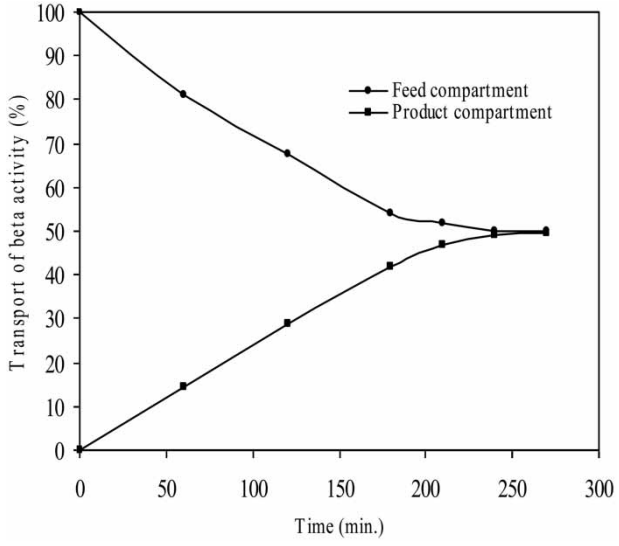

Figure 8. Transport of ^{90}Y in first stage using KSM-17 carrier in sequential mode.

yield for ^{90}Y separation in ~10 hrs. Results of the ^{90}Y transport in two stages using two single stage cells in sequential mode are depicted in Figs. 8 and 9.

Thus, in this improved generator system, the transport of ^{90}Y is achieved quantitatively in the first stage from a source phase at lower concentration of nitric acid (pH 2) to a higher concentration of nitric acid (3–4 M) through a PTFE membrane with a KSM-17 carrier. In the second stage, ^{90}Y is

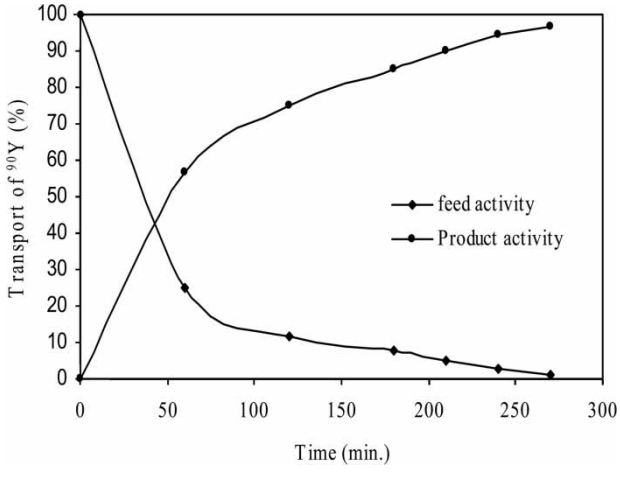

Figure 9. Transport of ^{90}Y in second stage using CMPO carrier in sequential mode.

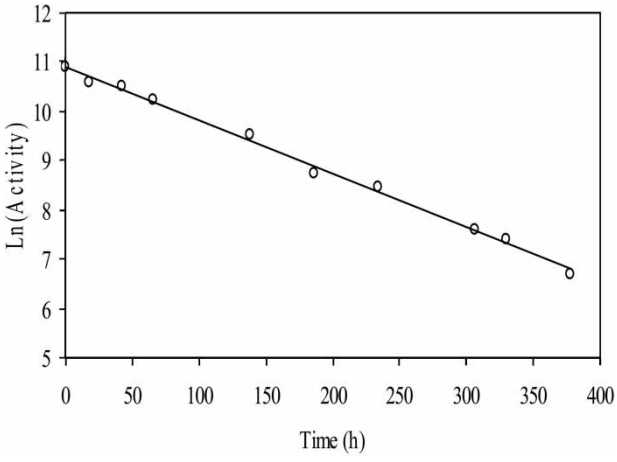

Figure 10. Decay curve of carrier-free ^{90}Y produced from two stage SLM.

transported through another PTFE membrane with CMPO carrier to the product compartment containing acetic acid. The second stage acts as a barrier that does not allow ^{90}Sr contamination in the product even if the generator system runs for prolonged period for ^{90}Y separation which was a drawback of the earlier single cell SLM (10).

Thus, by careful optimization of the configuration of the membrane cells, it is possible to obtain carrier free ^{90}Y in high yields free from ^{90}Sr, in a form suitable for direct labeling.

Quality of the ^{90}Y

The radiochemical purity of ^{90}Y is assayed by radiometric method. The β activity of the product plotted as a function of time (Fig. 10) indicated $T_{1/2}$ of $\sim$64 hours confirming the absence of radionuclide, other than ^{90}Y in the product. The initial β activity of about 10^5 cpm on the planchette is found to decay to the background activity after about 26 days.

CONCLUSIONS

A two stage supported liquid membrane based separation technique employing KSM-17 as carrier in the first stage and CMPO in the second stage is superior to the single stage SLM system developed earlier by us in terms of quality, yield, and chemical form. The second stage acts as a barrier for ^{90}Sr in case it accompanies the ^{90}Y in the intermediate compartment. The yield of the generator system is found to be $>95 \pm 3\%$ ($n = 4$)

in about 9–10 hours. The product is free from radiochemical impurities and is suitable for use in bio-medical applications. This technique is also amenable for scaling up.

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