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BHABHA ATOMIC RESEARCH CENTRE

**SEPARATION OF  $^{90}\text{Sr}$  FROM PUREX HIGH LEVEL WASTE AND  
DEVELOPMENT OF A  $^{90}\text{Sr}$ - $^{90}\text{Y}$  GENERATOR**

by

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ATOMIC ENERGY COMMISSION

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60	<i>Abstract :</i>	$^{90}\text{Y}$ ( $T_{1/2}=64.2$ h) finds several applications in nuclear medicine. It is formed from the decay of $^{90}\text{Sr}$ which has a long half-life of 28.8 years. $^{90}\text{Sr}$ can be used as a long-lasting source for the production of carrier-free $^{90}\text{Y}$ . $^{90}\text{Sr}$ itself is abundantly available in high level waste (HLW) of PUREX origin. The present studies deal with the separation of pure $^{90}\text{Sr}$ from HLW and the subsequent separation of $^{90}\text{Y}$ from $^{90}\text{Sr}$ . Actinides and some of the fission products like lanthanides, zirconium, molybdenum and cesium were first removed from the HLW using methods based on solvent extraction and ion-exchange studied earlier in our laboratory. The resulting waste solution was used as a feed for the present process. The separation of $^{90}\text{Sr}$ from HLW was based on radiochemical method which involved a repeated scavenging with ferric hydroxide followed by strontium carbonate precipitation. The separation of $^{90}\text{Y}$ from $^{90}\text{Sr}$ was achieved by membrane separation technique. A compact generator is developed for this separation using a commercially available polytetrafluoroethylene (PTFE) membrane, impregnated with indigenously synthesised 2-ethylhexyl 2-ethylhexyl phosphonic acid (KSM-17). Generator system overcomes the drawbacks associated with conventional solvent extraction and ion-exchange based generators. The product is in chloride form and is suitable for complexation studies. After gaining an operating experience of ~3 years in generating carrier-free $^{90}\text{Y}$ at 2 mCi level for initial studies in radiotherapeutic applications, the process was scaled up for the production of about 12 mCi of $^{90}\text{Y}$ to be used for animal studies before its application to patients. Radiochemical and chemical purity of the product was critically assayed by radiometry, ICP-AES, etc. The process is amenable for further scaling up.
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## SEPARATION OF $^{90}\text{Sr}$ FROM PUREX HIGH LEVEL WASTE AND DEVELOPMENT OF A $^{90}\text{Sr}$ - $^{90}\text{Y}$ GENERATOR

### ABSTRACT

$^{90}\text{Y}$  ( $T_{1/2} = 64.2$  h) finds several applications in nuclear medicine. It is formed from the decay of  $^{90}\text{Sr}$  which has a long half-life of 28.8 years.  $^{90}\text{Sr}$  can be used as a long-lasting source for the production of carrier-free  $^{90}\text{Y}$ .  $^{90}\text{Sr}$  itself is abundantly available in high level waste (HLW) of PUREX origin. The present studies deal with the separation of pure  $^{90}\text{Sr}$  from HLW and the subsequent separation of  $^{90}\text{Y}$  from  $^{90}\text{Sr}$ . Actinides and some of the fission products like lanthanides, zirconium, molybdenum and cesium were first removed from the HLW using methods based on solvent extraction and ion-exchange studied earlier in our laboratory. The resulting waste solution was used as a feed for the present process. The separation of  $^{90}\text{Sr}$  from HLW was based on radiochemical method which involved a repeated scavenging with ferric hydroxide followed by strontium carbonate precipitation. The separation of  $^{90}\text{Y}$  from  $^{90}\text{Sr}$  was achieved by membrane separation technique. A compact generator is developed for this separation using a commercially available polytetrafluoroethylene (PTFE) membrane, impregnated with indigenously synthesised 2-ethylhexyl 2-ethylhexyl phosphonic acid (KSM-17). Generator system overcomes the drawbacks associated with conventional solvent extraction and ion-exchange based generators. The product is in chloride form and is suitable for complexation studies. After gaining an operating experience of  $\sim 3$  years in generating carrier-free  $^{90}\text{Y}$  at 2 mCi level for initial studies in radiotherapeutic applications, the process was scaled up for the production of about 12 mCi of  $^{90}\text{Y}$  to be used for animal studies before its application to patients. Radiochemical and chemical purity of the product was critically assayed by radiometry, ICP-AES, etc. The process is amenable for further scaling up.

# SEPARATION OF $^{90}\text{Sr}$ FROM PUREX HIGH LEVEL WASTE AND DEVELOPMENT OF A $^{90}\text{Sr}$ - $^{90}\text{Y}$ GENERATOR

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## 1. INTRODUCTION

Yttrium-90 is identified as one of the important radionuclides for therapy in nuclear medicine. Suitability of this isotope is because of its short half-life ( $T_{1/2} = 64.2$  h), absence of gamma-ray-emissions and its high-energy beta emissions ( $E_{\text{max}} = 2.28$  Mev). Decay of  $^{90}\text{Y}$  generates a stable daughter product ( $^{90}\text{Zr}$ ). Its chemical properties are suitable for forming stable bindings with various chelating agents(1). It can be prepared by neutron-irradiation of yttrium metal or its oxide. But this method produces carrier-containing  $^{90}\text{Y}$ , which is a serious drawback for therapeutic applications.

Other source of  $^{90}\text{Y}$  is through the decay of  $^{90}\text{Sr}$ , the latter being abundantly present in high level waste (HLW) solutions of PUREX origin.  $^{90}\text{Sr}$  ( $T_{1/2} = 28$  y) is always in equilibrium with  $^{90}\text{Y}$  in HLW and hence  $^{90}\text{Sr}$  can form a perpetual source of  $^{90}\text{Y}$ , if separated from other constituents of HLW.  $^{90}\text{Sr}$  can be separated and obtained in a pure form using conventional radiochemical methods modified suitably(2).

Numerous methods for the separation of  $^{90}\text{Y}$  from  $^{90}\text{Sr}$  are available, which include precipitation, solvent extraction, ion exchange, extraction chromatography, etc.(3-7). Among these, ion-exchange has been the most commonly used method in the generator system for the preparation of carrier-free  $^{90}\text{Y}$ . The main drawback of this system is the leaching of  $^{90}\text{Sr}$  due to the associated radiation damage of the resin. Supported liquid membranes (SLM) based methods can be employed as an alternative technique for the separation of  $^{90}\text{Y}$  from  $^{90}\text{Sr}$ . This technique has many advantages over other conventional techniques, such as ease of operation, possibility of achieving high separation factors, economic use of extractants, etc.(8). Basic data for the SLM based technique can be gathered from conventional solvent extraction studies. Tri-n-butyl phosphate (TBP),  $\alpha$ -anilinobenzylphosphate, di-2-ethylhexyl phosphoric acid (HDEHP),

2-ethylhexyl 2-ethylhexyl phosphonic acid (KSM-17), etc. are the various extractants that can be used for the separation of  $^{90}\text{Y}$  from  $^{90}\text{Sr}$ (7,9,10).

Present work deals with the radiochemical separation of  $^{90}\text{Sr}$  from actinides, lanthanides and cesium depleted HLW of PUREX origin(11,12), followed by separation of carrier-free  $^{90}\text{Y}$  using KSM-17 based SLM technique. The radiochemical separation involved a scavenging with ferric hydroxide followed by strontium carbonate precipitation. The separated  $^{90}\text{Sr}$  was used as a feed for the production of  $^{90}\text{Y}$  using a compact generator system containing polytetrafluoroethylene (PTFE) membrane impregnated with KSM-17. The various experimental parameters like acidity of the feed, acidity of the receiver phase, concentration of strontium (added as carrier), time of operation, etc. which influence the transport of  $^{90}\text{Y}$  across SLM, were studied in detail. Radiochemical and chemical purity of the product was also assayed.

## 2. EXPERIMENTAL

### 2.1. Materials

All the reagents used were of analytical grade. Solvent extraction grade KSM-17, synthesised and characterised at U.R.E.D., B.A.R.C.(13) was used as an extractant and as a carrier in the solvent extraction and SLM experiments, respectively. Commercially available PTFE membranes were used in all the experiments. Membranes had a pore diameter and thickness of 0.45 and 160  $\mu\text{m}$ , respectively. The porosity of the membranes was about 84%. Membranes were impregnated by immersing them in KSM-17 for at least 12 hours before use and then washed with small quantity of water to remove the excess of extractant. The membrane cell (Fig. 1) was a two compartment permeation cell consisting of a feed compartment and a receiving compartment, each having a volume of 5 ml. The two compartments were separated by SLM having an effective membrane area of 1.13  $\text{cm}^2$ . The source and receiving phases were mechanically stirred to minimise the thickness of the aqueous diffusion layer.

### 2.2. Analysis

In all the experiments, gross beta activity was assayed by using a beta proportional counter standardised against a RaDE source and gross gamma activity using NaI(Tl) scintillation counter, standardised against a  $^{137}\text{Cs}$  source. Individual fission product gamma activities were assayed with a 62 c.c. HPGe detector coupled to a 4K multi-channel analyser. Chemical analysis

was carried out using inductively coupled plasma atomic emission spectroscopy (ICP-AES). In SLM studies, samples of 20 µl were taken from the feed as well receiving compartments periodically and analysed radiometrically using above detectors.

### 2.3. Methods

High level waste originated from the reprocessing of spent fuel is generally 3-4 M in HNO<sub>3</sub> and contains the fission products like <sup>90</sup>Sr-<sup>90</sup>Y, <sup>137</sup>Cs, <sup>144</sup>Ce, <sup>106</sup>Ru, <sup>95</sup>Zr-<sup>95</sup>Nb, actinides like U, Pu, Am, Np and corrosion products like Fe, Cr, Ni and Mn as well as process chemicals in the form of nitrate. The studies were carried out with HLW after depletion of actinides, lanthanides and cesium from it. Uranium, the major actinide constituent of HLW, was removed by solvent extraction using 30% TBP in n-dodecane. Plutonium and neptunium were also removed along with uranium(14). Minor actinides were separated using mixture containing 0.2 M octyl(phenyl)N,N-diisobutylcarbamoylmethylphosphine oxide (CMPO) and 1.2 M TBP in n-dodecane(11). The resulting HLW was lean in actinides, lanthanides and some of the fission products like zirconium. Cesium depletion was carried out by using ammonium phosphomolybdate(AMP)(12). This HLW, with depleted actinide, lanthanide and cesium, was used for all the further studies for the radiochemical separation of strontium and is referred hereafter simply as HLW. Some of the preliminary experiments were carried out using HLW diluted with 2 M nitric acid. In the experiments carried out to study the behaviour of strontium, the feed solutions were spiked with either <sup>90</sup>Sr or <sup>85+89</sup>Sr.

The radiochemical studies were carried out in centrifuge tubes, fabricated from borosilicate glass. Iron and strontium carriers were added in the form of ferric nitrate and strontium nitrate, respectively. After the addition of precipitant, the supernatant was separated from the precipitate by centrifugation followed by decantation. The detailed procedure of each of the experiments is presented in Results and Discussion.

In the preliminary solvent extraction studies, batch experiments were carried out by contacting a known volume of aqueous feed (generally 2 ml) containing required radionuclide in nitric acid medium of known acidity with equal volume of 20% KSM-17 in n-dodecane for 30 minutes. Organic and aqueous phases were separated, assayed for the activity and the distribution ratios were calculated.

For the SLM studies, 20 ml of the aqueous solutions containing the required radionuclide were prepared and the pH was adjusted to the required value. The pH adjustments were carried out by the addition of either 2 M nitric acid or 2 M NaOH. Five ml of these solutions were used as feed solutions in these studies.

### 3. RESULTS AND DISCUSSION

#### 3.1. Separation of $^{90}\text{Sr}$

Actinides were removed from PUREX HLW by solvent extraction using TBP and CMPO as extractants. This step removed lanthanides and some of the other fission products like zirconium along with the actinides. Cesium from this actinide-lean solution was removed by contacting it with AMP. The resulting HLW contained  $^{125}\text{Sb}$ ,  $^{106}\text{Ru}$  and traces of  $^{137}\text{Cs}$  in addition to  $^{90}\text{Sr}$ . The possibility of removal of these impurities was studied using radiochemical method.

Initial experiments were carried out to study the behaviour of  $^{90}\text{Sr}$  during the removal of impurities using iron as scavenger. Five ml of 1.0 M nitric acid solution containing  $^{90}\text{Sr}$  at tracer level were taken and 10 mg of  $\text{Fe}^{3+}$  were added as scavenger to this solution. Iron was precipitated as  $\text{Fe}(\text{OH})_3$  by addition of liquor ammonia. The radiometric analysis of the supernatant showed that more than 90% of the beta activity was carried down along with the ferric hydroxide precipitate. A similar experiment was carried out in presence of 10 mg of  $\text{Sr}^{2+}$  added as a carrier. The analysis of the supernatant showed that only ~20% of the beta activity was carried down with the precipitate under these conditions. These results show the necessity of addition of carrier strontium to prevent the co-precipitation of  $^{90}\text{Sr}$ .

In the next experiment, iron was precipitated from 5 ml of diluted HLW containing 10 mg each of  $\text{Fe}^{3+}$  and  $\text{Sr}^{2+}$  by addition of liquor ammonia. The precipitate was separated and washed with 2 ml of 1:1 ammonia. Washings were mixed with the supernatant and strontium was precipitated from this solution as strontium carbonate using 2% sodium carbonate solution. The precipitate was separated and dissolved in 5 ml of 2 M nitric acid. The analysis of this strontium solution (Sr-A) showed the removal of >99% of  $^{125}\text{Sb}$ ,  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  in this step (Table 1). The scavenging step was repeated with the addition of the same quantity (10 mg) of iron for further decontamination of the strontium from the impurities. This was followed by precipitation of strontium carbonate from the supernatant. The precipitate was dissolved in 5 ml of 2 M nitric

acid. The radiometric analysis of the solution (Sr-B) showed practically complete absence of  $^{125}\text{Sb}$ ,  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$ .

Further radiochemical separation studies were carried out using undiluted HLW. Since bio-medical applications demand for very high specific activity of  $^{90}\text{Y}$ , it is necessary to get a solution of  $^{90}\text{Sr}$  in high concentrations. Although this can be achieved through concentrating the waste through evaporation, further concentration of HLW resulted in crud formation/precipitation of some of the elements. As seen earlier, major amount of strontium is co-precipitated along with other radionuclides during the ferric hydroxide scavenging step, if no strontium carrier is added. Hence, a coprecipitation step with iron alone as carrier was used for pre-concentrating strontium in the present studies. The precipitate was dissolved in small volume of nitric acid. This formed the feed solution for strontium purification. The solution was spiked with iron as well as strontium. As in the radiochemical analysis, iron-carrier (precipitated as hydroxide) served as a scavenger for impurities and strontium-carrier maintained  $^{90}\text{Sr}$  in solution form.  $^{90}\text{Sr}$  was precipitated as strontium carbonate. Scavenging and precipitation steps were repeated to improve the decontamination of the strontium product from the impurities. The detailed procedure followed in these studies are given below and the results of the concerned analyses are presented in Table 2.

1. Five mg of iron (as ferric nitrate) were added to ~20 ml of actinide, lanthanide and cesium-lean HLW solution (~2 M) and precipitated as hydroxide using 6 ml of liquor ammonia. The precipitate, which contained the major fraction of  $^{90}\text{Sr}$  activity, was separated from the supernatant by centrifuging. The analysis of this supernatant (Supt-1) showed less than 10% of original gross  $\beta$  activity in it. Nearly 45% of  $^{125}\text{Sb}$ , 57% of  $^{106}\text{Ru}$  and 73% of  $^{137}\text{Cs}$  were left in the supernatant.
2. The activity left in the ammoniacal supernatant (Supt-1) was precipitated with second addition of 5 mg of  $\text{Fe}^{3+}$ . The supernatant (Supt-2) showed a further reduction in gross  $\beta$  activity to less than 5% of the original activity with the second addition of iron.  $^{125}\text{Sb}$ ,  $^{106}\text{Ru}$  and  $^{137}\text{Cs}$  activities were reduced to ~20%, ~42% and ~60%, respectively.
3. The precipitates from step 1 and 2, containing  $^{90}\text{Sr}$  and other fission products, were mixed and dissolved in 5 ml of 2 M nitric acid. This was followed by an addition of 25 mg of  $\text{Sr}^{2+}$  carrier. Total volume of the solution was 6 ml and it contained about 10 mg of iron.

4. Iron from the above solution was precipitated as  $\text{Fe}(\text{OH})_3$  using 6 ml of liquor ammonia. (Due to the presence of strontium carrier, major fraction of  $^{90}\text{Sr}$  remained in the supernatant.) Supernatant of this scavenging step was separated. The precipitate was discarded after giving a wash with 3 ml of 1:1 ammonia solution. Supernatant and washings were mixed (Supt-3, total volume ~15 ml) and analysed for gross  $\beta$  and individual fission product activities. Results indicate that more than 90% of the fission products present in the original feed were removed during this scavenging step.
5. Strontium from the supernatant of step 3 (Supt-3) was precipitated as strontium carbonate using 7 ml of 2% sodium carbonate solution. The analysis of the supernatant (Supt-4) showed the total gross  $\beta$  activity to be <1% of that originally present in the feed. This step also shows good decontamination of the product ( $^{90}\text{Sr}$ ) with respect to the individual fission products.
6. The strontium carbonate precipitate was dissolved in ~4 ml of 1 M nitric acid. This solution was subjected to repeated scavenging and carbonate precipitation steps (3 times) for further decontamination from fission products as described in step 4 onwards. Ten mg of iron were added in each scavenging step. No strontium was required to be added as the solution had already ~25 mg of strontium in it.

First repetition of scavenging and carbonate precipitation generated Supt-5 and Supt-6, whereas Supt-7 and Supt-8 were generated in the next repetition, respectively. Supt-7, generated in final scavenging step, showed only small quantity of  $^{106}\text{Ru}$  (0.005% of originally present in the feed), with practically no detectable activity of  $^{125}\text{Sb}$  and  $^{137}\text{Cs}$ .  $^{106}\text{Ru}$  was not detected in the supernatant from the subsequent carbonate precipitation.

7. Final strontium carbonate precipitate was washed with 3 ml of 2% sodium carbonate and dissolved in minimum volume of 1.0 M nitric acid. This solution (Prdt-Sr) containing  $^{90}\text{Sr}$  was made up to 6 ml with 0.1 M nitric acid.

Radiochemical purity of the above strontium product (Prdt-Sr) was assayed (Table 3). The solution contained 5.24 mCi/ml of gross  $\beta$  activity immediately after separation (with no  $^{90}\text{Y}$  in it). Although no  $^{125}\text{Sb}$  and  $^{137}\text{Cs}$  was detected in the product, it contained ~44 nCi of  $^{106}\text{Ru}$ /ml. This corresponds to 8.4 nCi of  $^{106}\text{Ru}$ /mCi of  $^{90}\text{Sr}$ . (During the subsequent separation of  $^{90}\text{Y}$  from  $^{90}\text{Sr}$  by SLM technique,  $^{90}\text{Y}$  product did not show the presence of  $^{106}\text{Ru}$ .) The growth of  $^{90}\text{Y}$  was followed through beta counting using proportional counter for the quantification of  $^{90}\text{Y}$  activity.

The scheme used for the radiochemical separation of  $^{90}\text{Sr}$  from HLW is summarised in Fig.2.

### 3.2. Separation of $^{90}\text{Y}$

#### 3.2.1. Solvent extraction studies:

Preliminary experiments were carried out to compare the extraction behaviour of  $^{90}\text{Sr}$  and  $^{90}\text{Y}$  from low nitric acid medium using 20% KSM-17 in n-dodecane as extractant. The acidity of the aqueous feed was varied from 0.5 M to pH 5. The results of these studies are presented in Table 4. In general, the extraction of yttrium was much higher compared to that of strontium and increased substantially with decrease in acidity. Practically no extraction of  $\text{Sr}^{2+}$  was observed from 0.5 M and pH 1 solutions. At higher pH, the distribution ratios were of the order of  $10^{-3}$ . As expected for the acidic extractants, extraction increased substantially with decrease in acidity. The separation factors ( $D_{\text{Y}}/D_{\text{Sr}}$ ) were of the order of  $10^4$  and above for all the feed acidities.

Radiochemical separation of  $^{90}\text{Sr}$  from HLW uses carrier-strontium. Hence, extraction behaviour of yttrium was studied from pH 1 solution in presence of varying concentration of strontium (up to 10 mg/ml) using 20% KSM-17 as extractant. The extraction of yttrium increased with increase in strontium concentration in the aqueous phase (Table 5). Distribution ratio for  $^{90}\text{Y}$  increased from  $\sim 0.4$  in absence of strontium to  $\sim 9$  in presence of 10 mg/ml of strontium in the aqueous phase. This increase in extraction with increase in strontium nitrate concentration may be because of salting-out effect of nitrate ions. Probably KSM-17 behaves as a neutral extractant under these conditions.

#### 3.2.2. Transport studies:

The studies carried out from solutions with feed acidities of pH 1 and pH 2 using 0.5 M HCl in the receiver phase showed a transport of  $\sim 30\%$  in 6 hours in both cases (Table 6). The transport was marginally higher from a feed solution of pH 2. Similar studies were carried out from feed solution with an acidity of pH 1 containing 5 mg/ml of strontium. The results showed that the transport is improved from  $\sim 30\%$  to  $\sim 40\%$ . These results are concurrent with those from solvent extraction studies reported above.

The effect of concentration of hydrochloric acid in the receiving phase on the transport of  $^{90}\text{Y}$  was studied in the next set of experiments. The results of these experiments are presented in

**Table 7.** The feed solution used in these experiments contained 5 mg/ml of strontium and had an acidity of pH 1. The rate of transport of  $^{90}\text{Y}$  increased substantially from ~40% to ~80% with an increase in the acidity of the receiving phase from 0.5 M to 3.0 M.

Since the radiochemical purity of  $^{90}\text{Y}$  is very important in bio-medical applications, it is necessary to study the co-transport of  $^{90}\text{Sr}$ . The transport of strontium was studied from feed solutions containing 3 mg/ml of strontium and having different acidities (pH 1, 2 and 3). In these experiments carried out using  $^{85+89}\text{Sr}$  as tracer, 0.5 M hydrochloric acid was used as a receiving phase. The results of these experiments are presented in Table 8. The results showed that the transport of strontium increased with decreasing feed acidity. Practically no transport of strontium was observed even after 72 hours with a feed solution having an acidity of pH 1. In the case of feed solution with an acidity of pH 2, about 0.45% of strontium was detected in the receiving phase after 48 hours, which increased to ~1% in 72 hours. The transport was considerably higher with a feed solution of pH 3. About 0.26% of strontium was transported within first 3 hours and it reached to 3.65% within 6 hours. The transport was as high as 11.37% after 72 hours.

Transport of strontium was also studied from a feed solution with an acidity of pH 1 and containing 3 mg/ml of strontium spiked with  $^{85+89}\text{Sr}$ . No strontium was detected upto 72 hours, in this experiment carried out using 0.5 M hydrochloric acid as a receiving phase. After 96 hours, a marginal transport of about 0.0003% was observed. In the experiments carried out with a feed containing 5 mg/ml of strontium and having same acidity of pH 1, no transport of strontium was observed up to 6 hours when 1 M hydrochloric acid was used as a receiving phase. A marginal transport of ~0.0008% was observed after 12 hours. Under similar conditions, even in the presence of higher concentration of 10 mg/ml of strontium in the feed, receiving compartment did not show any detectable strontium activity after 6 hours. It can be concluded that the generator system can be operated for 6 hours under above conditions to produce  $^{90}\text{Y}$  product free from strontium activity. These results were confirmed by ICP-AES analysis.

The above studies show that the transport of yttrium increases with increasing acidity of the receiving phase and that of strontium decreases with increase in the acidity of the feed. A feed with an acidity of pH 1 (0.1 M) and a receiving phase of 1.0 M hydrochloric acid results in satisfactory transport of about 60% of yttrium within 6 hours with practically no detectable transport of strontium. The transport of yttrium was reduced to ~37% when the acidity of the

feed was increased from 0.1 M to 0.5 M. These results indicate that a feed acidity of pH 1 and a receiving phase of 1 M hydrochloric acid can be used for the separation of  $^{90}\text{Y}$  from  $^{90}\text{Sr}$ .

About 30 mCi of  $^{90}\text{Sr}$  separated from HLW as reported above in the radiochemical separation studies was used as a feed for the generator system for the separation of  $^{90}\text{Y}$  using KSM-17 based SLM technique. Using this generator system, ~12 mCi of clinical grade  $^{90}\text{Y}$  were separated for bio-medical applications. The transport of  $^{90}\text{Y}$  was ~40% within first 3 hours and increased to ~43% within next 3 hours. The radiometric assay did not show any gamma-emitting radionuclides in the  $^{90}\text{Y}$  product. Decay of the product was followed by beta counting and showed an half-life of 64.17 hours (Figure 3). The residual activity was found to be matching with that of the background. Chemical purity of the product was also assayed by ICP-AES. No impurities were detected in the product.

A smaller version of the generator was regularly used to separate and supply about 2 mCi of  $^{90}\text{Y}$  to Radiopharmaceuticals Division for complexation studies, for almost 3 years on weekly basis.

#### 4. CONCLUSION

HLW generated during the reprocessing of spent fuel can be used as a source of  $^{90}\text{Sr}$ . Highly radiotoxic alpha-emitters (actinides) and fission products can be separated from HLW solution using solvent extraction and ion-exchange techniques. Solvent extraction using TBP and CMPO as extractants removes practically all actinides, lanthanide and some of the fission products like zirconium, molybdenum, etc. Inorganic ion-exchangers like AMP can be used for the removal of cesium from the HLW. The actinide, lanthanide and cesium-lean HLW forms a suitable feed for  $^{90}\text{Sr}$ , which can be purified by radiochemical methods involving repeated iron scavenging followed by strontium carbonate precipitation. The product  $^{90}\text{Sr}$  constitutes an ideal feed for generation of carrier-free  $^{90}\text{Y}$ , which can be repeatedly milked after allowing for its growth. Using KSM-17 based SLM technique, carrier-free  $^{90}\text{Y}$  can be separated from  $^{90}\text{Sr}$ . The strontium carrier added during the radiochemical separation step enhances the transport of  $^{90}\text{Y}$  during membrane based separation. A yield of ~40% is reached within only 3 hours with this generator system. The product is free from radiochemical impurities and can be used for bio-medical applications.

## 5. ACNOWLEDGEMENT

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**TABLE 1****Radiochemical Separation of Strontium from HLW: Preliminary Experiment****Experimental Conditions**

Feed: Diluted Actinides, Lanthanides and Cesium depleted HLW

Volume = 5 ml

Quantity of Fe added = 10 mg in each step.

Quantity of Sr added = 10 mg.

Precipitant Used: Liquor ammonia.

Sample	Gross $\beta$ (mCi/l)	$^{125}\text{Sb}$ (mCi/l)	$^{106}\text{Ru}$ (mCi/l)	$^{137}\text{Cs}$ (mCi/l)
Feed	522.00	7.90	159.60	6.35
Sr - A	157.53	0.050	0.201	0.029
Sr - B	122.66	ND	ND	ND

Sr - A: Sr product after first  $\text{Fe}^{3+}$  scavenging followed by carbonate precipitationSr - B: Sr product after second  $\text{Fe}^{3+}$  scavenging followed by carbonate precipitation

ND: not detected

**TABLE 2****Distribution of Activity During Radiochemical Separation of Strontium in HLW****Experimental Conditions**

Feed: Actinides, lanthanides and cesium depleted HLW

Volume = 20 ml

Total quantity of Fe added = 10 mg

Quantity of Sr added = 10 mg.

Precipitants used: Liquor ammonia and 2% Na<sub>2</sub>CO<sub>3</sub>

Sample	% Distribution *				
	Gross $\beta$	Gross $\gamma$	<sup>125</sup> Sb	<sup>106</sup> Ru	<sup>137</sup> Cs
Supt-1	8.55	30.32	45.33	56.56	73.24
Supt -2	4.68	19.68	19.88	42.11	60.32
Supt -3	25.90	8.95	6.21	10.31	8.88
Supt -4	0.75	3.60	3.99	7.54	7.45
Supt -5	17.55	2.31	0.35	0.45	0.47
Supt -6	0.13	0.14	0.24	0.42	0.53
Supt -7	16.89	1.69	0	0.005	0
Supt -8	0.35	0.05	0	0	0
Prdt-Sr	15.62	1.59	0	0.016	0

\* With respect to alpha and cesium lean HLW

**TABLE 3**  
**Radiochemical Purity of <sup>90</sup>Sr Product**

Sample	Gross $\beta$	Impurities		
		<sup>125</sup> Sb	<sup>106</sup> Ru	<sup>137</sup> Cs
Feed (20 ml, Actinides, lanthanides and cesium depleted HLW)	11.18 (mCi/ml)	33.98 ( $\mu$ Ci/ml)	92.77 ( $\mu$ Ci/ml)	22.05 ( $\mu$ Ci/ml)
Product (6 ml, in 0.1 M HNO <sub>3</sub> )	5.24 (mCi/ml)	ND	44.1 (nCi/ml)	ND

ND: not detected

**TABLE 4****Extraction of Strontium and Yttrium from Nitric Acid Medium  
Using 20% KSM-17 in n-Dodecane**

Experimental conditions

Phase ratio = 1:1

Volume = 2 ml

Contact Time = 30 minutes

Feed (Acidity/pH)	$D_Y$	$D_{Sr}$	Separation Factor ( $D_Y / D_{Sr}$ )
0.5 M	0.35	No Extraction	Very high
pH 1	2.66	No Extraction	Very high
pH 2	116.57	$2.7 \times 10^{-3}$	$4.3 \times 10^4$
pH 3	126.40	$4.4 \times 10^{-3}$	$2.9 \times 10^4$
pH 4	140.36	$6.6 \times 10^{-3}$	$2.1 \times 10^4$
pH 5	209.86	$9.9 \times 10^{-3}$	$2.1 \times 10^4$

**TABLE 5****Extraction of Yttrium from Nitric Acid Medium in Presence of Strontium  
Using 20% Ksm-17 in n-Dodecane****Experimental conditions**

Feed acidity = pH 1

Volume = 2 ml

Phase ratio = 1:1

Contact Time = 30 minutes

Conc. of Sr (mg/ml)	Total NO <sub>3</sub> <sup>-</sup> (M)	D <sub>Y</sub>
0	0.100	0.44
2.0	0.146	0.89
5.0	0.214	1.81
7.0	0.260	5.66
10.0	0.328	8.89

**TABLE 6****Transport of  $^{90}\text{Y}$  Through KSM-17 Supported Liquid Membrane**

Experimental conditions

Feed:  $^{90}\text{Y}$  (tracer) in nitric acid medium, volume = 5 ml

Strippant: 0.5 M HCl, volume = 5 ml

Time (min)	% Transport	
	Feed: pH = 1	Feed: pH = 2
15	0.32	1.08
30	1.83	2.06
60	4.35	4.76
120	8.66	9.85
180	11.23	12.70
240	19.60	22.70
300	24.47	25.00
360	28.92	33.14

**TABLE 7****Transport of  $^{90}\text{Y}$  Through KSM-17 Supported Liquid Membrane in Presence of Strontium with Different Concentrations of Hydrochloric Acid as Strippant****Experimental conditions**

Feed:  $^{90}\text{Y}$  (tracer) in nitric acid medium at pH 1  
containing 5 mg/ml of Sr, volume = 5 ml

Strippant: HCl, volume = 5 ml

Time (min)	$^{90}\text{Y}$ Transport (%)			
	Strippant: 0.5 HCl	Strippant: 1 M HCl	Strippant: 2 M HCl	Strippant: 3 M HCl
15	1.38	1.79	2.05	2.74
30	3.15	4.68	6.85	7.02
60	6.98	9.31	14.60	21.38
120	14.40	19.88	32.97	41.12
180	21.92	25.32	48.78	55.72
240	32.87	41.20	61.44	67.96
300	38.04	56.10	69.71	77.15
360	40.40	61.04	74.56	81.87

**TABLE 8****Transport of  $^{85+89}\text{Sr}$  Through KSM-17 Supported Liquid Membrane**

Experimental conditions

Feed:  $^{85+89}\text{Sr}$  in nitric acid medium, volume = 5 ml

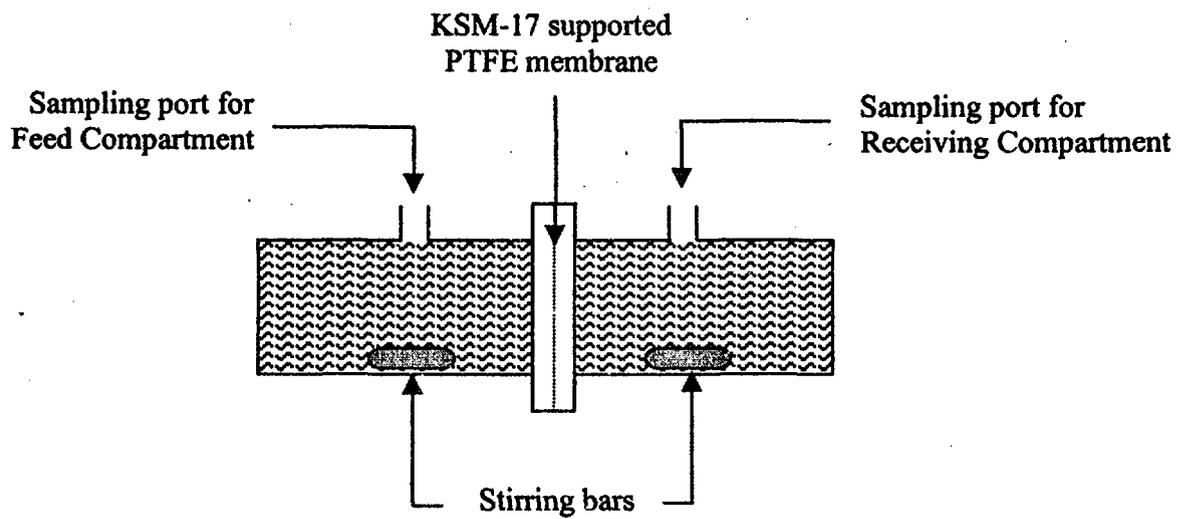
Strippant: 0.5 M HCl, volume = 5 ml

Time	% Transport		
	Feed: pH = 1	Feed: pH = 2	Feed: pH = 3
15 min	ND	ND	ND
30 min	ND	ND	ND
1 hr	ND	ND	ND
2 hr	ND	ND	ND
3 hr	ND	ND	0.26
4 hr	ND	ND	1.54
5 hr	ND	ND	2.06
6 hr	ND	ND	3.65
24 hr	ND	ND	6.77
48 hr	ND	0.45	-
72 hr	ND	1.04	11.37

ND = Not detected

**TABLE 9****Transport of Strontium Through KSM-17 Based Supported Liquid Membrane**

Experimental conditions	Time	% Transport
Feed: 3 mg of Sr/ml, pH = 1 Receiving phase: 0.5 M HCl	6 hrs	ND
	72 hrs	ND
	96 hrs	0.0003
Feed: 5 mg of Sr/ml, pH = 1 Receiving phase: 1.0 M HCl	6 hrs	ND
	12 hrs	0.0008
Feed: 10 mg of Sr/ml, pH = 1 Receiving phase: 1.0 M HCl	6 hrs	ND



**Fig.1. Schematic Diagram of Membrane Cell**

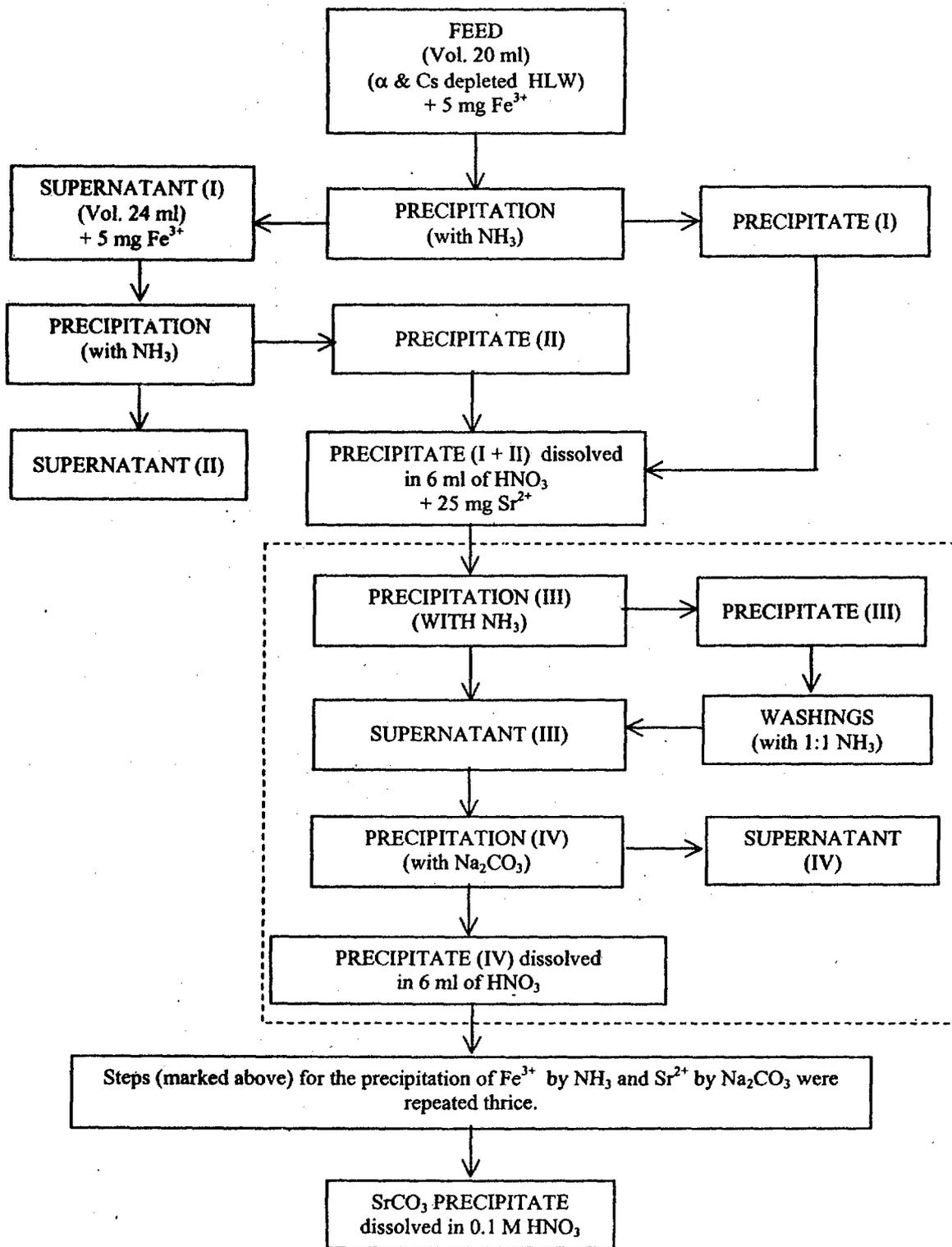
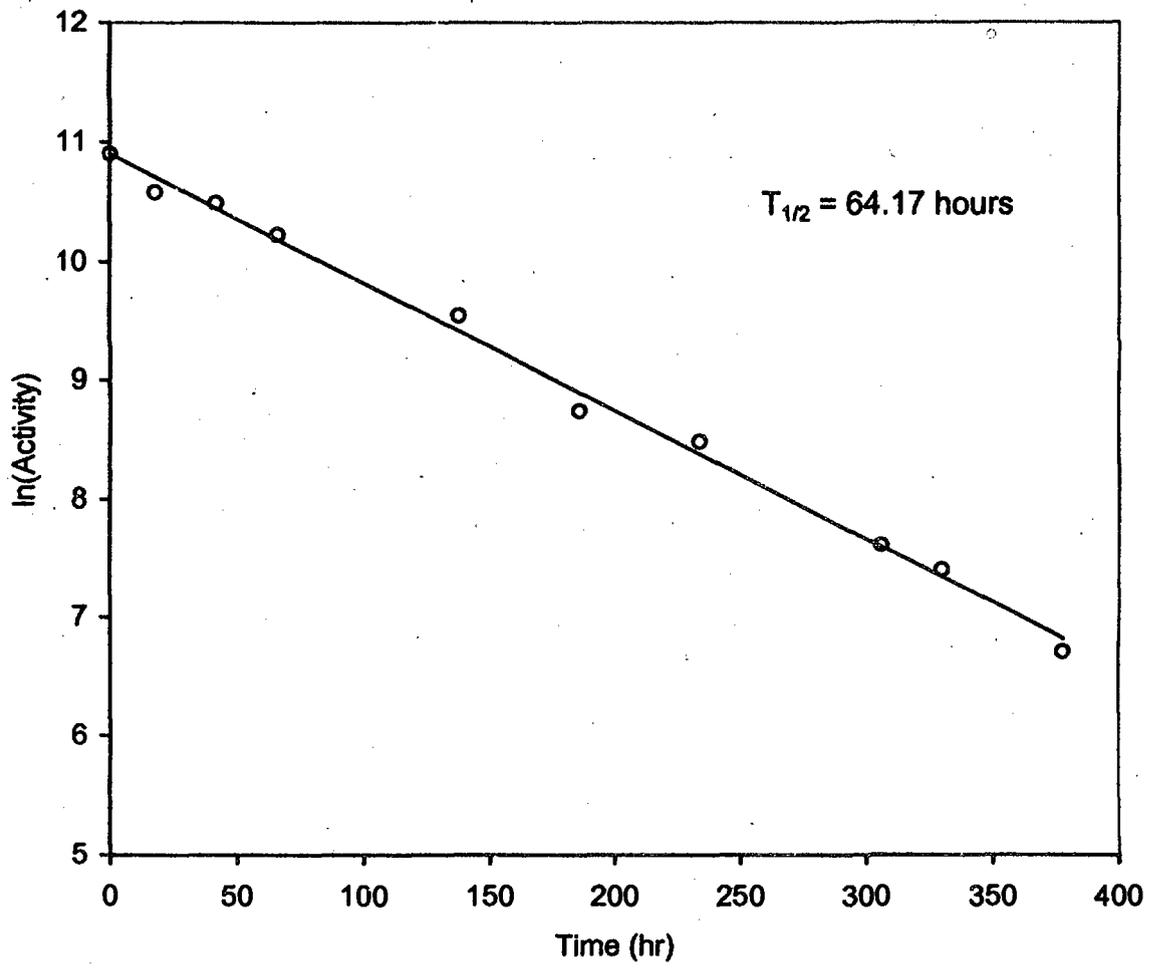


Fig.2. Scheme for Separation of <sup>90</sup>Sr from HLW



**Fig.3. Decay Curve of carrier-free  $^{90}\text{Y}$  Produced from  $^{90}\text{Sr}$ - $^{90}\text{Y}$  Generator**

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