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# Separation of Carrier-Free $^{90}$Y from High Level Waste by Extraction Chromatographic Technique Using 2-Ethylhexyl-2-ethylhexyl Phosphonic Acid (KSM-17)

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## ABSTRACT

An extraction chromatographic technique has been developed for the separation of carrier-free $^{90}$Y from the $^{90}$Sr present in the high level waste (HLW) of the Purex process. When a Purex HLW solution in 2–3 M $HNO_3$ is passed through a CMPO-Chromosorb-102 (CAC) column, all the trivalent, tetravalent, and hexavalent ions are sorbed. The effluent from this experiment, after adjusting the pH to 2 with NaOH, was passed through a 2-ethylhexyl-2-ethylhexyl phosphonic acid (KSM-17)-Chromosorb-102 (KSMC) extraction chromatographic column where only $^{90}$Y was sorbed. All the other ions ($^{90}$Sr, $^{137}$Cs, $^{125}$Sb, $^{106}$Ru, $^{106}$Rh, etc.) were washed off with dilute $HNO_3$ (pH 2), and carrier-free $^{90}$Y was eluted with 0.5 M $HNO_3$. This technique can yield $^{90}$Y in mCi levels in pure form for medical applications. The $^{90}$Sr can be used repeatedly after allowing for $^{90}$Y buildup.

## INTRODUCTION

$^{90}$Y is one of the most useful radioisotopes in nuclear medicine. It is a pure β emitter with no associated gamma and ends up in a stable daughter $^{90}$Zr. In the recent past many studies have been reported on its use in radioimmuno therapy of cancers (1–7). It can be prepared by irradiation of yttrium metal or oxide (8), but the resulting product has low specific activity and this is a serious disadvantage in therapeutic applications. The other major source of $^{90}$Y is

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the parent fission product $^{90}$Sr present in the high level waste (HLW) originating from the Purex process (9). This source, once separated, can repeatedly produce large amounts of carrier-free $^{90}$Y, is less costly, and can serve as a long-term source because of the 28 year half-life of the parent $^{90}$Sr (10). But the main problems are associated with the isolation of pure $^{90}$Sr from the HLW, which is a highly complex, radiotoxic, and hazardous waste.

Many reports are available on the separation of $^{90}$Y from $^{90}$Sr, including precipitation (11), solvent extraction (12, 13), and ion-exchange chromatography (14). But most of these techniques suffer from specific problems like corrosion due to acid, organic extracting agent contamination, and incomplete separation from $^{90}$Sr. Ion-exchange chromatography is the most frequently used $^{90}$Y generation technique. A serious disadvantage for clinical use of an ion-exchange generator system is the high level of $^{90}$Sr breakthrough due to radiation damage. A generator system consisting of a column packed with inorganic adsorbants gave only 60% $^{90}$Y yield (15). Preparation of carrier-free $^{90}$Y from $^{90}$Sr by extraction chromatography using di-(2-ethylhexyl)phosphoric acid supported on Teflon grains for medical applications has been reported (16), but this method employs corrosive 8 N HCl as the eluting medium for $^{90}$Y and the feed does not contain any other impurities.

Octyl(phenyl)-*N*,*N*-diisobutylcarbamoylmethylphosphine oxide (CMPO) mixed with tributyl phosphate (TBP) or supported on an inert matrix is capable of sorbing all alpha emitting nuclides and fission products in their hexa-, tetra-, and trivalent forms, leaving behind a waste containing mainly the β emitters $^{90}$Sr and $^{137}$Cs with trace amounts of $^{106}$Ru and $^{125}$Sb (17). The present paper reports an approach for the separation of carrier-free $^{90}$Y using 2-ethylhexyl-2-ethylhexyl phosphonic acid (KSM-17, equivalent to PC-88A) from actinide and Cs-depleted HLW obtained after CMPO (17) and zeolite treatment, respectively.

## EXPERIMENTAL

### Materials

The inert support, Chromosorb 102 (styrene divinylbenzene polymer, Johns Manville, USA), of 100–120 mesh size was given an acetone wash and air-dried. KSM-17 was synthesized at this Centre and was purified by a procedure reported in the literature (18).

### Preparation of KSMC

Impregnation of Chromosorb-102 with KSM-17 was carried out (in a 1:3 w/w ratio of Chromosorb·102 and KSM-17) in 100% methanol (19). The impregnated resin (KSMC) was washed with 20% methanol and air-dried.

## Feed Solution

The actinide–lanthanide and Cs depleted HLW had an acidity of around 0.5 M $HNO_3$. A suitable aliquot of this solution was diluted and adjusted to pH 2 with 1 M NaOH. This solution was used as the feed for extraction chromatography. The gamma spectrum of the feed is given in Fig. 1.

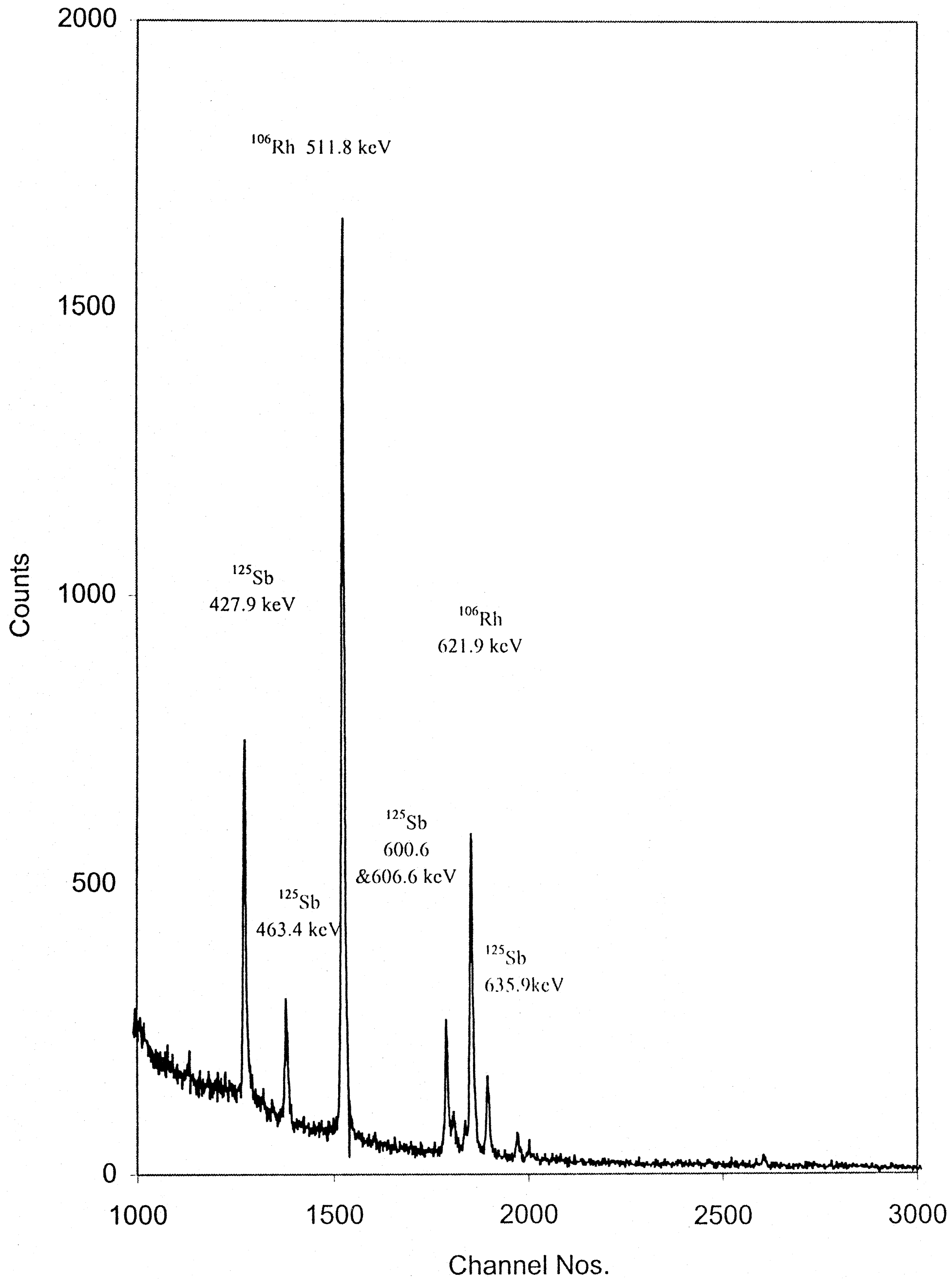

FIG. 1 Gamma spectrum of the feed.

## Wash Solution

An aqueous solution adjusted to pH 2 with nitric acid was used as the wash solution for the removal of weakly sorbed and occluded activities of undesirable impurities from the extraction chromatographic column.

## Column Studies

A column (ϕ0.4 × 9 cm) containing 0.5 g of KSMC was loaded with 5 mL of feed at a flow rate of 5 bed volumes per hour. The loaded column was washed until the activity of the effluent almost reached the background level. Elution of the sorbed $^{90}$Y was carried out with 0.5 M $HNO_3$. Fractions of the eluted product were assayed for alpha, beta, and gamma activities.

## Sample Analysis

Suitable aliquots of the samples drawn at various stages were assayed for gross beta using a β proportional counter standardized against a RaDE source and gross gamma using a NaI(Tl) scintillation counter standardized against *a* $^{137}$Cs source. Individual fission product gamma activities were assayed with a 62-cc HPGe detector coupled to a 4K multichannel analyzer.

The experimental parameters and the feed assay are given in Table 1.

# RESULTS AND DISCUSSION

The alpha emitting nuclides of Pu, U, Np, Am, and Cm and all trivalent lanthanides from Purex HLW were removed quantitatively by successive

TABLE 1

| | |
|---|---|
| Weight of KSMC | 0.5 g |
| Column dimension | ϕ 0.4 × 9 cm |
| Feed volume | 5 mL |
| Wash solution | 0.01 M $HNO_3$ |
| Eluant | 0.5 M $HNO_3$ |
| Flow rate | 5 bed volume/hour |
| Feed composition: | |
| $H^+$ | 0.01 M $HNO_3$ |
| Gross α | Below detection limit |
| Gross β | 786.71 mCi/L |
| Gross γ | 9.59 mCi/L |
| $^{106}$Ru-$^{106}$Rh | 1.75 mCi/L |
| $^{125}$Sb | 0.35 mCi/L |

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contacts with 30% TBP and CMPO, leaving behind Sr, Cs, Pd, Tc, Ru, Sb, and a few other metal ions in the effluent (17). This effluent had an acidity around 2 M. This was diluted to bring down the acidity to 0.5 M. The resulting solution was passed through a column containing zeolite which removed the Cs selectively, leaving behind Sr and other ions in the effluent. After a few weeks, $^{90}$Y ($T_{1/2}$-64.2 hours) formed by the beta decay of $^{90}$Sr reached secular equilibrium with its parent and formed the ideal feed for $^{90}$Y recovery. Solvent extraction and extraction chromatograpic studies using KSM-17 showed practically no uptake for almost all mono- and divalent ions present in this effluent at pH 2. $Y^{3+}$ could thus be selectively sorbed onto KSMC from a feed solution at pH 2, leaving behind all other mono- and divalent ions.

Estimation of the $^{90}$Sr in the feed was carried out using a radiochemical procedure reported in the literature (20). As the feed is an aged solution, the activity of $^{90}$Y is the same as that of $^{90}$Sr. In an earlier study reported (19) from our laboratory using KSM-17-based extraction chromatography, it was shown that trivalent nuclides can be firmly held on the column without any breakthrough as long as the loading and washing solution pH is maintained at 2. The gamma spectrum (Fig. 1) shows the peaks of $^{106}$Rh and $^{125}$Sb in the feed, and indicates $^{106}$Ru-$^{106}$Rh and $^{125}$Sb as the probable contaminants along with $^{90}$Sr in the $^{90}$Y product. The gamma spectrum of the effluent from the column (Fig. 2) shows all the peaks present in the feed that are not retained in the column. During the washing of the loaded column with 0.01 M $HNO_3$, all the unsorbed and weakly sorbed nuclides are removed as is evident from the decreasing beta, gamma activities of the effluent. Once the beta value of the effluent reaches the background level, $^{90}$Y can be eluted out with 0.5 M $HNO_3$. At this acidity the beta activity of the effluent was found to increase and all the $^{90}$Y free from other gamma emitters could be eluted out, as can be seen from the gamma spectrum of the product (Fig. 3). More than 90% of the loaded $^{90}$Y activity could be eluted with 30 mL of the eluent. A concentrated product can be obtained in 10 mL volume if 1 M acid is used as eluent in place of 0.5 M $HNO_3$.

The log beta activity of the product has been plotted as a function of time (Fig. 4) and a $T_{1/2} \approx 64$ hours is obtained, confirming the presence of only $^{90}$Y. The initial β activity of about $10^5$ cpm on the planchette was found to decay to the background activity after a month. Radiochemical analysis of a single complete lot of $^{90}$Y product after a month failed to give any β counts above the background activity. Thus the decontamination factor achieved from $^{90}$Sr by this technique is well over $10^5$. The $^{90}$Sr source can be used repeatedly after allowing a time period for the buildup of $^{90}$Y.

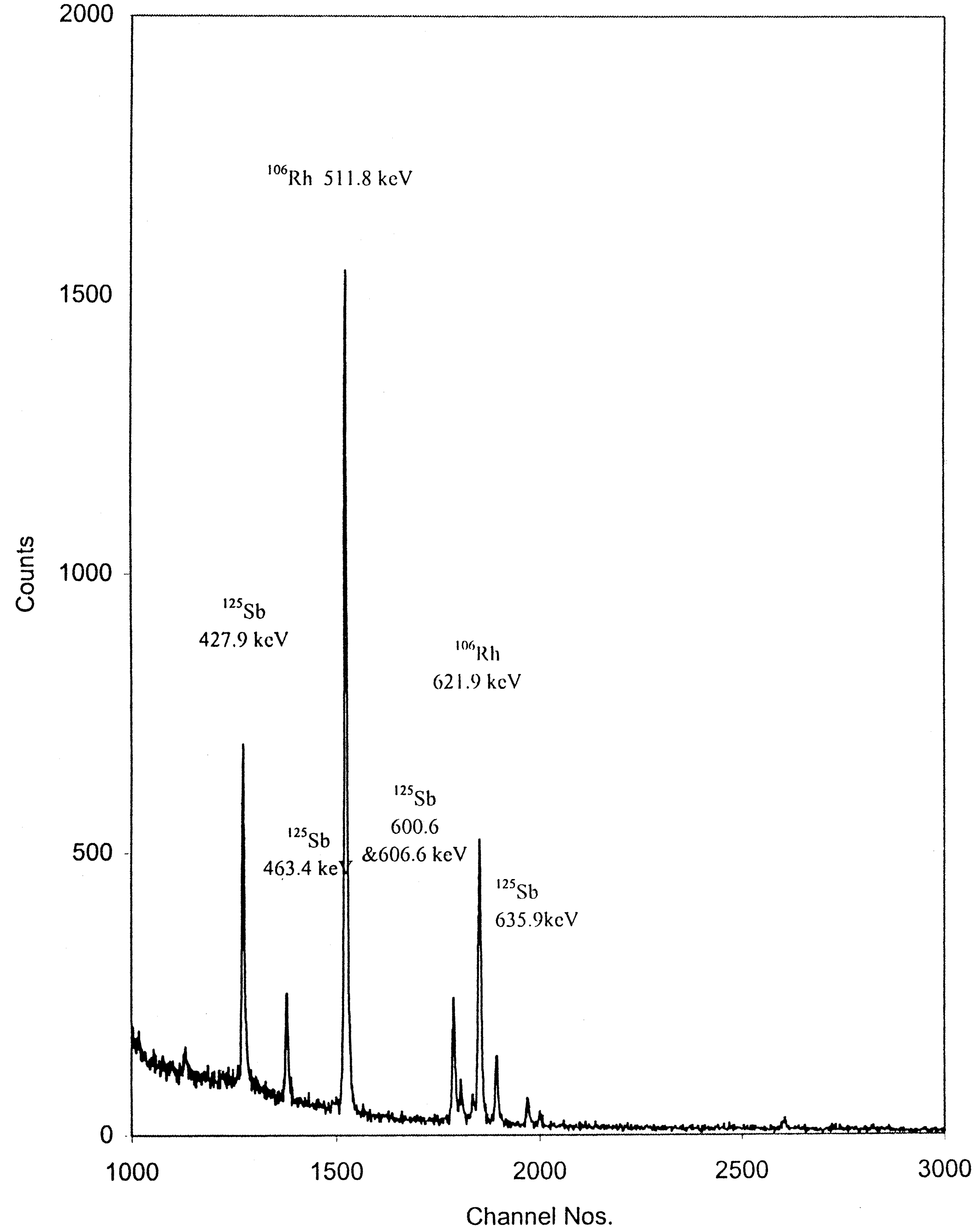

FIG. 2 Gamma spectrum of the effluent.

## CONCLUSION

$^{90}$Y in pure form for medical applications can be separated from $^{90}$Sr recovered from the Purex HLW by this simple technique. The first two steps for the respective removal of alpha and $^{137}$Cs activities need to be carried out only

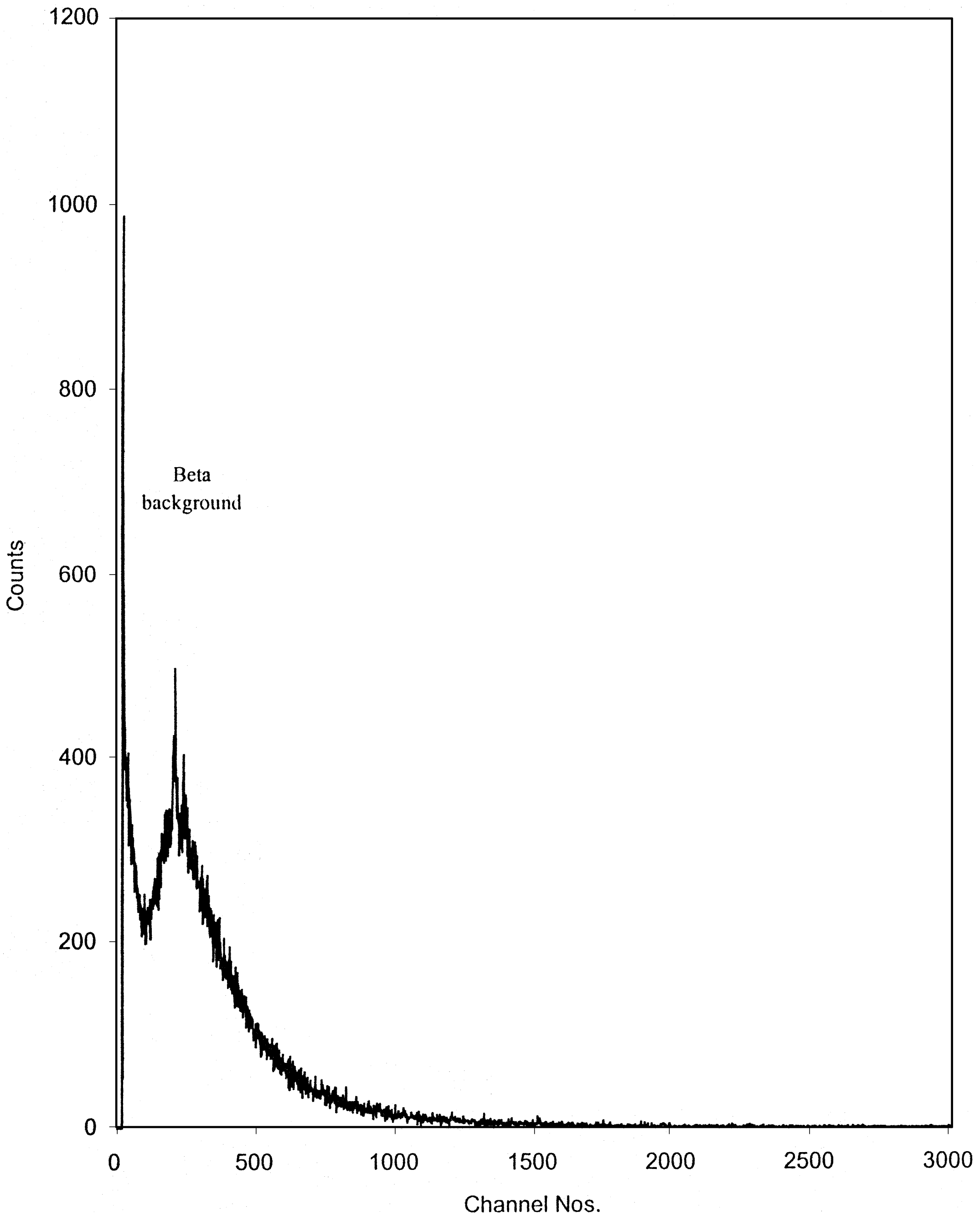

FIG. 3 Gamma spectrum of the $^{90}$Y product.

once. The effluent from the KSMC column can be processed at frequent intervals for $^{90}$Y recovery since the half-life of the parent $^{90}$Sr is 28 years. By using this technique $^{90}$Y in mCi levels has been separated. Since the eluted sample is in dilute nitric acid, it can be easily converted to any desired form. Based

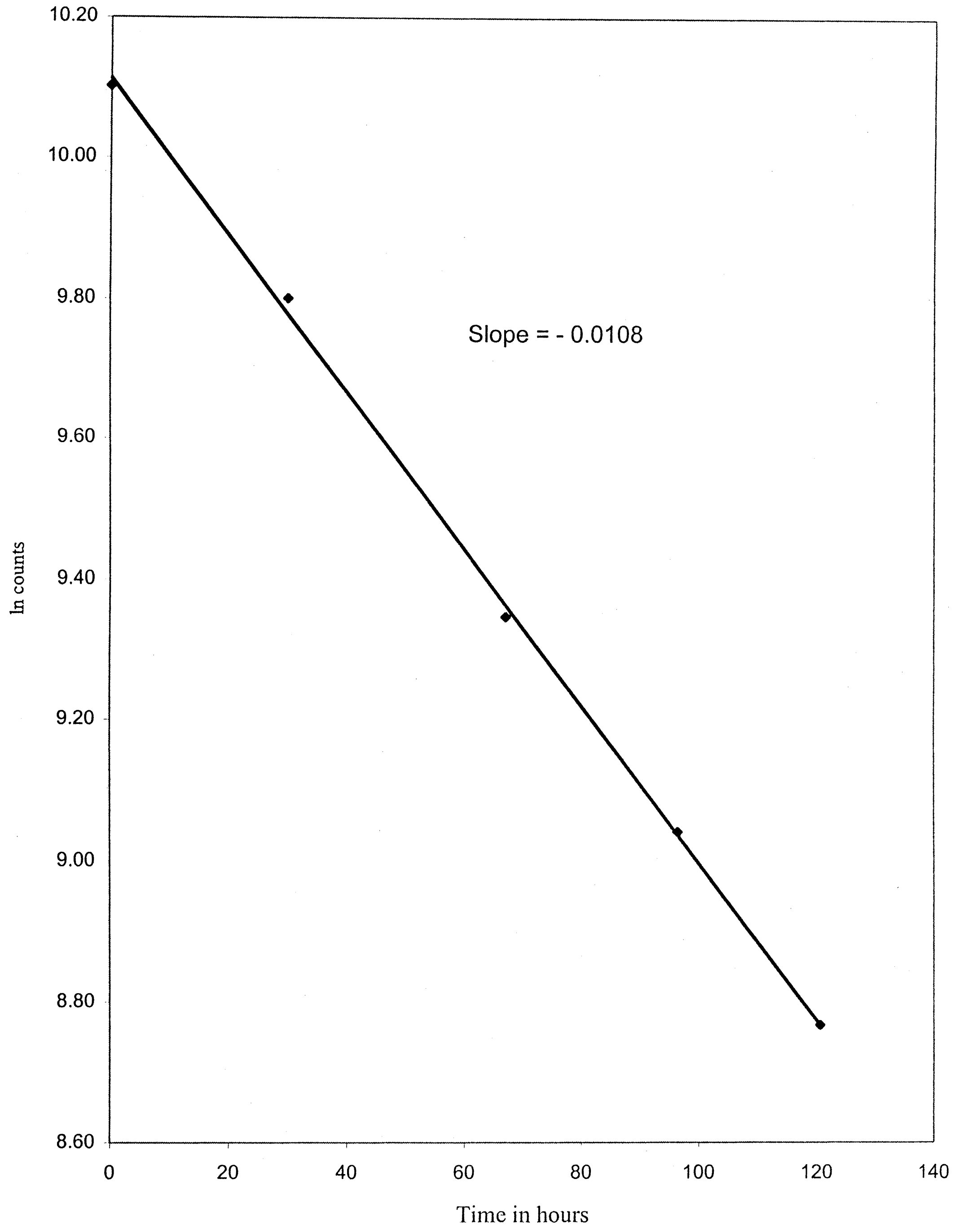

FIG. 4 Plot of log(natural) beta counts vs time of $^{90}$Y.

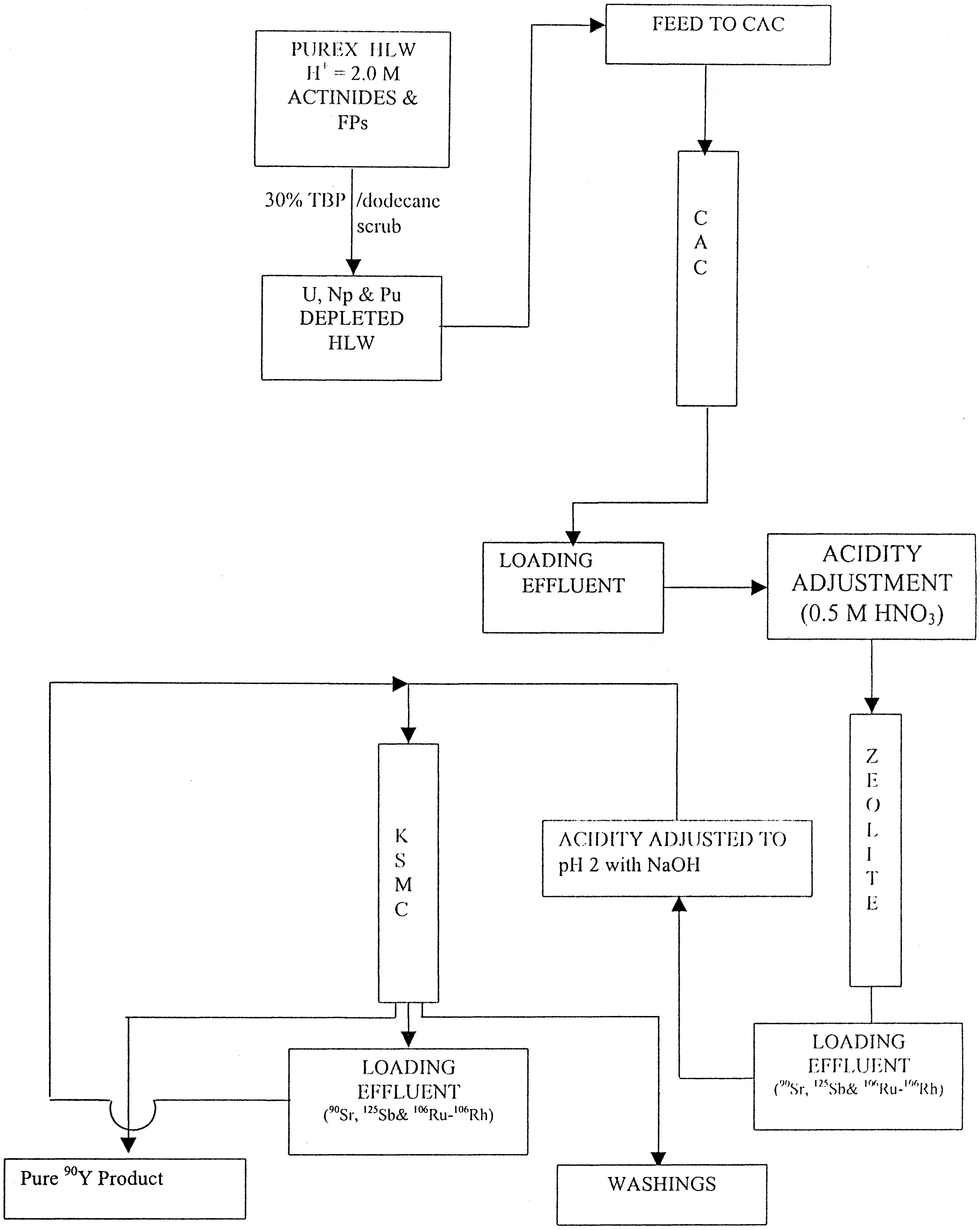

FIG. 5 Schematic diagram for the separation of $^{90}$Y from Purex HLW.

on the above studies, a scheme (Fig. 5) has been proposed for the separation of carrier-free $^{90}$Y from Purex HLW.

## REFERENCES

1. D. J. Hanatowich, F. Virzi, and P. W. Doherty, *J. Nucl. Med.*, *26*, 503 (1985).
2. S. E. Order, J. L. Klein, P. K. Leichner, J. Francke, C. Lollo, and D. J. Carlo, *Int. J. Radiat. Oncol. Biol. Phys.*, *12*, 277 (1986).
3. M. Roselli, J. Schlom, O. A. Gansow, A. Raubitschek, S. Mirzadeh, M. W. Brechbiel, and D. Colcher, *J. Nucl. Med.*, *30*, 672 (1989).
4. S. J. Mather, D. M. Tolley, and G. W. White, *Eur. J. Nucl. Med.*, *15*, 307 (1989).
5. B. A. Morton, B. G. Beatly, A. D. Mison, P. M. Wanek, and J. D. Beatly, *Cancer Res. (Suppl.)*, *50*, 1008S (1990).
6. J. M. Estaban, D. M. Hyams, B. C. Beatty, B. Merchant, and J. D. Beatly, *Cancer Res.*, *50*, 989S (1990).
7. L. C. Washburn, T. T. Sun, Y. C. C. Lee, B. L. Byrd, E. C. Holloway, J. E. Crook, J. B. Stubbs, M. G. Stabin, M. W. Brechbiel, O. A. Gansow, and Z. Steplewski, *Nucl. Med. Biol.*, *18*, 313 (1991).
8. *Radioisotope Production and Quality Control Technical Report* (Series No. 128), IAEA, Vienna, 1971, p. 520.
9. D. N. Sunderman and C. W. Townley, *Nuclear Science Series* (NAS-NS-3201), National Academy of Science–National Research Council, Washington, DC, 1960.
10. H. Chember, *Introduction to Health Physics*, 2nd ed., Pergamon Press, Oxford, 1983, p. 187.
11. R. Salutsky and M. Kirly, *Anal Chem.*, *27*, 576 (1955).
12. J. S. Wike, C. E. Guyer, D. W. Ramey, and B. P. Phillips, *Appl. Radiat. Isot.*, *41*, 861 (1990).
13. T. W. Lee and G. Ting, *Isotopenpraxis*, *27*, 269 (1991).
14. W. J. Skraba, H. Arino, and H. H. Kramer, *Int. J. Appl. Radiat. Isot.*, *29*, 91 (1978).
15. C. L. Hsieh, C. Z. Ma, and Y. K. Lu, *Nucl. Med. Mag. (China)*, *12*(2), 184 (1991).
16. B. T. Hsieh, G. T. Hsieh, and L. H. Shen, *Appl. Radiat. Isot.*, *44*, 1473 (1993).
17. J. N. Mathur, M. S. Murali, R. H. Iyer, A. Ramanujam, P. S. Dhami, V. Gopalakrishnan, M. K. Rao, L. P. Badheka, and A. Banerji, *Nucl. Technol.*, *109*, 216 (1995).
18. J. A. Partridge and R. C. Jenson, *J. Inorg. Nucl. Chem.*, *31*, 2587 (1969).
19. A. Ramanujam, P. V. Achuthan, P. S. Dhami, V. Gopalakrishnan, R. Kannan, and J. N. Mathur, *Solv. Extr. Ion Exch.*, *13*, 301 (1995).
20. D. N. Sunderman and C. W. Townley, *The Radiochemistry of Barium, Calcium and Strontium*, NAS–NS-3010, Washington, DC, 1960.

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