



AUSTRALIAN ATOMIC ENERGY COMMISSION
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FISSION PRODUCT SEPARATION FROM BERYLLIUM BY ION EXCHANGE

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ABSTRACT

An ion-exchange process has been developed on the laboratory scale for the separation of low level fission product activity from beryllium in aqueous nitrate solution. A 97 per cent. recovery of beryllium with gross beta and gamma decontamination factors of 10^4 and 2×10^3 respectively has been demonstrated. The process is flexible and should be applicable to both sulphate and chloride media.

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1. INTRODUCTION

When beryllia is used as moderator and matrix in a dispersion-type reactor fuel, its recovery may be necessary because of its relatively high cost. The beryllia will be contaminated with fission products due to recoil and diffusion processes in the reactor, and will become further contaminated in the separation processes used to recover the fissile and fertile components.

The H.T.G.C. reactor system under study at the A.A.E.C. Research Establishment uses such a beryllia based fuel. A simple method for the recovery and decontamination of beryllium from fission products was required, assuming the prior removal and recovery of the actinide elements. Since the fuel cycle for the proposed reactor had not been defined when this work commenced, it was assumed that the fuel would be treated by the conventional solvent extraction processes based on nitric acid dissolution followed by tributyl phosphate extraction of the actinides. In such a system the beryllium remains with the fission products in the waste nitrate solution leaving the solvent extraction process.

A considerable number of laboratory procedures have been developed for the separation and purification of beryllium from various metals (Moore 1960), but as yet no process has been developed for the separation and purification of beryllium from concentrated aqueous solutions contaminated with fission products.

Blanco, Higgins, and Kibbey (1956) successfully separated and decontaminated aluminium from process waste solutions contaminated with fission products, using a combination of scavenging precipitation and ion exchange. The present work was undertaken to demonstrate on the laboratory scale, the use of ion exchange to separate and decontaminate beryllium from a similar waste solution.

2. EXPERIMENTAL

2.1 Reagents

(a) Resins

The resins used for the cation-exchange studies were Zeo-Karb 225, 20-50 mesh and 50-100 mesh, 4-5% divinylbenzene (D.V.B.), 8% D.V.B., and 12% D.V.B., a sulphonic acid type cation-exchange resin, and Zeo-Karb 215, 20-50 mesh and 8% D.V.B., a sulphonated phenol-formaldehyde type resin. For the anion-exchange study, De-Acidite FF, a strong base anion exchanger, 100-200 mesh, and containing 3-5% divinylbenzene, was used.

(b) Radioactive Tracers

The elements chosen for preliminary investigations were representative of the major fission products existing after a cooling period of one year from a conventional power reactor. The following isotopes were purchased from the Radiochemical Centre, Amersham, England, and used as tracers: Cs-137, Sr-85, Ce-144, Zr-95, and Ru-106. The tracers were obtained as or converted into the nitrate form.

The mixed fission products used were separated from uranyl nitrate solution obtained from the dissolution of urania (Isaacs, unpublished work, 1964).

Beryllium concentrations were measured by scintillation counting on samples of the liquid phase, using Be-7 as a tracer. This was also purchased from the Radiochemical Centre.

(c) Chemicals

All chemical reagents were of analytical grade.

2.2 Techniques

(a) Batch equilibration

Batch equilibrium techniques were used in the preliminary studies. Equilibrations were

carried out at $25^{\circ} \pm 0.1^{\circ} \text{C}$ in an apparatus described by Bishop, de Bruin, and Temple (1960). Polyethylene containers were used in place of glass to minimize the loss of beryllium and fission products by sorption.

(b) Column operation

Beryllium elution data were obtained by saturating various size columns with beryllium and measuring the number of bed volumes of the eluent necessary to remove the beryllium.

Both glass and polyethylene columns were used throughout this study. The use of polyethylene columns was necessary when solutions containing fluoride ions were investigated. Glass wool was employed as a bed support for the glass columns, and a fibre-glass mesh enclosing polyethylene shavings was used as a support for the polyethylene columns. Various column sizes, 48.0 cm x 0.60 cm, 48.0 cm x 1.29 cm, and 11.0 cm x 1.29 cm, were used in this investigation, but the 48.0 cm x 0.6 cm polyethylene column was used for most of the work.

Normal column operations were performed, care being taken to allow for the volume above the column, the interstitial volume, and the volume between the bed support and column tip.

(c) Radiometric analysis

Total gamma activity was determined by scintillation counting on samples of the liquid phase. For simultaneous beta and gamma measurements, aliquots of the solution were evaporated on stainless steel planchets and counted on a beta-gamma anti-coincidence counter. Column operations involving mixed fission products were followed using a gamma spectrometer (256-channel analyzer, RCL model 20617).

3. RESULTS

The experimental work began with a series of preliminary experiments to determine the choice of the resin, the eluent, and the major operating conditions. Batch equilibrations were used as a fairly rapid sorting test, but column testing was necessary to confirm that chosen conditions were optimum. The results are given in Tables 1 - 15 and Figures 1 - 7.

4. DISCUSSION OF RESULTS

4.1 The Choice of a Resin

Initial investigations were made with the sulphonated polystyrene cation-exchange resin Zeo-Karb 225 and the sulphonated phenol-formaldehyde cation-exchange resin Zeo-Karb 215. As the latter was available only in 20 - 50 mesh range particle size all comparative tests were made with Zeo-Karb 225 in the same size range. The examination of Zeo-Karb 215 was prompted by its reported superior radiation stability (Costachel and Furmicha 1958) - a loss of only 1 per cent. of its capacity per watt hour of radiation adsorbed (per gram of oven dry resin), compared with a loss of 10 - 20 per cent. of their capacity by sulphonated polystyrene type resins. However, the capacity of the phenolic type resin is approximately one third that of Zeo-Karb 225 since the phenolic group rarely participates in ion exchange below a pH value of 10.0.

Experiments showed that Zeo-Karb 215 was subject to continuous colour throwing in dilute nitric acid. In strong nitric acid chemical attack of the resin was very noticeable. In contrast Zeo-Karb 225 was chemically resistant to the medium.

Further comparative tests of the resins were made by measuring the sorption of each of the major fission products as a function of beryllium concentration. Table 1 and Figures 1 and 2 summarize the results from these measurements. The slopes of the sorption functions for Cs-I, Sr-II, and Ce-III on Zeo-Karb 215 are close to the respective values of 1, 2, and 3 predicted by ion-exchange theory. However, with Zeo-Karb 225 as the adsorbing medium, Cs-I departed from an ideal ion-exchange behaviour in beryllium concentrations greater than 1.0M, and Sr-II and Ce-III deviated in beryllium concentrations greater than 0.2M. Neither Zr-95 or Ru-106 obeyed the exchange equivalence for an ion-exchange mechanism, which is not surprising in view of their more complex aqueous chemistry.

Zeo-Karb 225 was chosen for use in further development of the process because of its greater chemical stability in nitric acid and its stronger sorption of the alkaline and rare earth elements.

For column operation under near-equilibrium conditions it is essential to use as fine a resin particle size as possible, but the degree of fines is limited by the need for practical flow rates.

The basis of the process under investigation is the sorption of fission products high on the ion-exchange column during loading of the column with beryllium in order to prevent cross contamination of the beryllium when it is selectively eluted from the column. Thus a chromatographic type column (48.0 cm x 0.6 cm) was chosen for the major part of the study. To avoid inefficient separation through channelling, 20-25 mesh resin was rejected in column separation experiments in favour of 50-100 mesh resin. Impractical flow rates were obtained when 100-200 mesh resin was used.

Since resin selectivity is a function of ionic size and concentration of active sites on the resin, a decrease in resin porosity should increase selectivity differences. Therefore fission product sorption by the hydrogen form of Zeo-Karb 225 was determined as a function of the degree of resin cross-linking. This study was performed in 0.1M beryllium nitrate solution and the results are contained in Table 2 and Figure 3. Comparison of columns 3 and 5 in Table 2 shows the influence of the form of the resin on fission product sorption. Figure 3 shows that an increase in resin porosity decreased fission product sorption for Cs-137, Sr-85, and Ce-144, but neither Zr-95 nor Ru-106 was affected by this change.

Considering only those fission products that form simple cations in solution, the distribution coefficients in Tables 1 and 3 show that Cs-137 will adsorb lowest on an ion-exchange column. This radionuclide therefore dictates the magnitude of many of the column parameters.

With an increase in the degree of resin cross-linking there is an increasing danger of cracked beads caused by changes in electrolyte concentration. This produces fines and decreases column flow rates. Thus, while there was an increase in the sorption of Cs-137 with an increasing divinylbenzene content from 8 per cent. to 12 per cent., the increase was insufficient to offset the disadvantages caused by column choking.

Thus 50-100 mesh, 8% D.V.B., Zeo-Karb 225 appeared the most suitable resin and was chosen to test the chemical feasibility of separating and decontaminating beryllium from beryllium nitrate solutions containing fission products.

4.2 Flow Rate

In general, low flow rates produce sharper breakthrough curves and higher breakthrough capacity for a sulphonated resin than do high flow rates. The effect of flow rate depends largely on particle diffusion and hence the size of the resin particles and their degree of cross-linking. For a porous resin with a low degree of cross-linking, the flow rate has little influence, whereas for the highly cross-linked resin this factor is significant, especially if the particles are coarse.

The results of ion-exchange separations performed in other laboratories indicated that a flow rate of 1 millilitre per minute per square centimetre would be adequate for the chosen resin.

4.3 Sorption Cycle

4.3.1 Beryllium concentration

For the maximum separation of fission products from beryllium the beryllium concentration in the feed must be low so that fission product sorption on the column is high as predicted by the mass ion theory of ion exchange. This is clearly seen when Table 1 is examined. To enhance Cs-137 sorption 0.1M beryllium solutions were used in the majority of the ion-exchange separation runs. Optimum conditions for fission product separation were obtained for this feed solution before investigations were extended to a study of the effects of increasing the beryllium concentration in the feed while maintaining wash and elution cycle conditions constant.

Measurements were made of the effects of increasing the beryllium concentration on beryllium loading characteristics and Cs-137 sorption. Increasing the beryllium concentration from 0.1M to 0.2M and 0.5M reduced the breakthrough capacity by 4 per cent. and 6 per cent. respectively. No Cs-137 was detected in the effluent even after the column was loaded to saturation.

Subsequent column tests (Table 3) showed that little change occurred in the purity of the beryllium product with the increase in the beryllium concentration of the feed. Gamma spectra of the resulting products possessed no Cs-137 peak, showing that for the chromatographic column all the Cs-137 was adsorbed from each of the feeds.

Before a process can be based on a high beryllium feed concentration a balance must be struck between the beryllium concentration in the elution cycle effluent and the feed. If the recovered beryllium concentration is less than that of the feed, then it is of no advantage to work with the higher feed concentration. However, an ion-exchange process for beryllium recovery from a solution with concentration greater than 0.1M beryllium is feasible (Runs 26 - 27 and 35 - 36, Table 3).

4.3.2 Acidity of the feed

The influence of the feed acidity on fission product sorption was determined from a series of batch equilibrium studies. The examination was made in 0.2M beryllium nitrate solution, the pH of which was adjusted to between 0.5 to 3.0 with nitric acid and ammonium hydroxide. Both Zeo-Karb 225 and Zeo-Karb 215 were used in this investigation. A study of the latter resin was prompted by the possibility that the phenolic group may undergo reaction with increasing pH. The results in Table 4 and Figure 4 show that fission product sorption by Zeo-Karb 225 remained reasonably constant in the pH region 1.0 - 3.0 except for Ru-106. Increasing the pH favoured Ru-106 adsorption. Since ruthenium normally exists in several forms in solution, increasing the pH must alter the equilibrium in favour of the extractable species. Fission product sorption by Zeo-Karb 215 (summarized in Table 4 and Figure 5) showed a similar pattern for Zr-95. Zeo-Karb 215 possesses a far higher affinity for Zr-95. Below a pH value of 1.0 and except for Zr-95, there was a general reduction in fission product sorption.

Breakthrough capacity tests with Zeo-Karb 225, 50 - 100 mesh, 8% D.V.B., ion-exchange resin and 0.1M beryllium nitrate solutions showed that increasing the acidity of the solution decreased the breakthrough capacity. For a solution at a pH value of 2.10, 145 milligrams of beryllium were adsorbed (per 14.4 ml of resin); an increase of acidity to a pH value of 1.0 decreased the capacity to 127 milligrams of beryllium. Despite this decrease in beryllium loading, all separation studies were made at a pH value of 1.0. The choice of the acidity was finally dictated by the correct use of complexing ligand in the feed.

4.3.3 Complexing agents in the feed

The radionuclides involved in the separation studies may be classified into two groups. The first includes those fission products which exist exclusively as simple cations and must be sorbed high on the ion-exchange column. This is achieved by suitable choice of ion-exchange operating conditions. The second group comprises those fission products which do not form simple cations in solution or which co-exist in several ionic forms. Zirconium and ruthenium fall into this latter group. It is these two fission products which limit the ultimate separation of many processes.

Complexing agents were investigated for the conversion of zirconium and ruthenium into suitable forms which might pass through or be strongly sorbed on the cation exchanger during the sorption cycle.

(a) Thiourea

The use of the urea analogue, thiourea, for the conversion of ruthenium into cationic complexes and subsequent sorption on a carboxylic type resin has been reported by Prohaska (1958). It was decided to test this method using a sulphonated resin.

Batch equilibrium techniques were used to examine the sorption of Ru-106 by both Zeo-Karb 225 and Zeo-Karb 215 in the presence of thiourea, as a function of pH. The results are shown in Table 5. Solution preparation was based on Prohaska's method.

The sorption of Ru-106 by both cation exchangers was enhanced by the presence of thiourea. An increase in pH favoured the sorption by Zeo-Karb 225 but did not alter the affinity of Ru-106 for Zeo-Karb 215.

Fission product sorption studies were then performed using Prohaska's method. Table 6 shows that no significant change occurred with Cs-137, Sr-85, or Ce-144. Zr-95 sorption increased slightly, but hydrolysis effects could account for this.

Subsequent column testing of thiourea in the presence of solutions containing mixed fission products was not so encouraging (Table 7). Beta and gamma decontamination factors were low, but gamma spectrometer analyses indicated that the chief cause was poor Zr-95 separation. The addition of a silica gel column failed to produce a significant increase in Zr-95 decontamination. The gamma spectra revealed that Ru-106 sorption had increased, but not to the extent predicted from batch equilibrium results.

The successful application of oxalic acid to the separation of both Ru-106 and Zr-95 from beryllium, discussed fully below, resulted in the abandonment of the thiourea studies.

(b) Oxalic acid

Other workers (Bruce 1956) have studied aqueous soluble complexing agents for decreasing zirconium extractability in solvent extraction processes. Oxalic acid was the most promising reagent (Ferguson 1956).

Earlier work in this laboratory by de Bruin, Kairaitis, and Temple (1962) demonstrated that beryllium forms neutral and anionic complexes with oxalic acid. However, the strength of these complexes is such that significant complexing occurs only above a pH value of 2.0. A pH value of 1.0 was chosen for all beryllium sorption studies in the presence of oxalic acid.

Batch equilibrium studies were carried out on solutions of tracer amounts of Be-7 and Zr-95 at a pH value of 1.0 in the presence and absence of a large excess of oxalic acid (0.1M). The results (Table 8) show that oxalic acid had little effect on beryllium sorption, but reduced the zirconium to an insignificant level. The strong interaction of Zr-95 with Zeo-Karb 215 recorded in Table 1 was tested in the presence of oxalic acid as a final confirmation of the formation of the zirconium-oxalate complex. The negligible interaction of macro-concentrations of beryllium (0.1M) with 0.1M oxalic acid at pH of 1.0 was substantiated by similar loading characteristics obtained in 0.1M nitric acid and 0.1M oxalic acid.

Studies of the sorption of individual fission products from 0.1M beryllium nitrate solution (pH = 1.0) containing 0.1M oxalic acid showed that Cs-137 and Sr-85 remained unaffected by oxalic acid but the sorption of both Ce-144 and Ru-106 was reduced (Table 9). However the Ce-144 sorption was still sufficiently large to permit its separation. The reduction of the Ru-106 sorption to a negligible level showed that oxalic acid can be effective in removing the majority of Ru-106 and Zr-95 activity from the beryllium.

The maximum concentration of oxalic acid that can be tolerated in a 0.1M beryllium feed solution was determined by measuring the breakthrough volume for beryllium at various oxalic acid concentrations. Results (Figure 6) indicate 0.1M oxalic acid is the limiting concentration. No investigation has been made of the optimum concentration necessary for the maximum separation of Zr-95 and Ru-106 from beryllium. Final measurement of this will require high level activity runs in a high activity facility.

In Runs 12-15 of Table 7 no oxalic acid was included in the feed and a gamma decontamination factor of 2×10^2 was obtained. However, Runs 16-19 and 21-25 in Table 10 show that the inclusion of oxalic acid resulted in gamma D.F. values of 10^3 .

(c) EDTA

Based on the work of Aggett (1962), the possibility of substituting EDTA for oxalic acid was investigated. Aggett showed that EDTA effectively complexes a large proportion of the fission products if the solution is refluxed for 4 hours. This complex action is also pH dependent, the higher the pH the more universal EDTA is as a complexing agent. However, beryllium will complex with EDTA at a pH value greater than 4.5 (Merrill, Honda and Arnold 1960), so that a separation was performed at a pH value of 4.0. The addition of Na^+ for alkali adjustments of pH and the use of the sodium form of EDTA introduced the complication that Cs-137 might elute with the beryllium in the elution cycle. An auxiliary column of zirconium phosphate was therefore used to absorb the Cs-137 (Fardy, unpublished work, 1965), before the addition of EDTA. Furthermore, the cation-exchange resin was saturated to only 85 per cent. of its practical capacity to enable good fission product adsorption.

The results of the separation are summarized in the flowsheet of Figure 7. The beryllium yield was 97 per cent. with an overall gamma decontamination factor of 2.5×10^3 , which compared favourably with the figures obtained when oxalic acid was used. No attempt was made to examine the parameters associated with the EDTA system but the results warrant a closer examination in the future.

Ideally the method for obtaining pure beryllium is to adsorb the fission products on an ion-exchange resin while the beryllium passes through the column. However, the number, the various oxidation states, and the variable nature (ions and radio-colloids) of the fission products do not permit this simple process. This is supported by the results in Figure 7 where the 'single pass' method for separating fission products from beryllium was tested. After the solution was passed through the zirconium phosphate column and the EDTA added, the fission products were sorbed on the nitrate form of the anion exchanger De-Acidite FF, 100-200 mesh, 3-5% D.V.B. The inefficiency of this type of process is reflected in the poor overall gamma D.F. value of 1.2

(d) Beryllium Saturation of the Column

Ion-exchange column studies of fission product solutions revealed that although oxalic acid complexed most of the zirconium, a small amount of the element remained on the column and was eluted with the beryllium. Similar behaviour was noted for Ru-106 despite the use of either oxalic acid or thiourea. To minimise the residual Zr-95 and Ru-106 on the column, columns were saturated to beryllium breakthrough capacity. Column loading experiments for 0.1M beryllium feed and Zeo-Karb 225, 50-100 mesh, 8% D.V.B., ion-exchange resin revealed little difference between the breakthrough and practical loading volumes with the chromatographic column 48.0 cm x 0.60 cm (see Table 11). Thus the extension of column saturation to practical loading has little effect on the overall process.

4.4 Wash Cycle

In early separation studies 0.02M oxalic acid was used in the wash cycle in order to flush any residual Zr-95 activity from the column. The successful application of oxalic acid in the feed solution in later studies emphasised the need for a wash cycle containing oxalic acid. A study was made of the maximum concentration of this acid that could be tolerated by the beryllium after presetting the total volume of this wash cycle at two resin bed volumes. The results (Table 11) showed that 0.05M oxalic acid was a suitable concentration. Increasing the column saturation with beryllium from breakthrough to practical loading approximately doubled the concentration of beryllium eluted by 0.05M oxalic acid.

Gamma spectra from Run 16 and onwards showed that the inclusion of 0.1M oxalic acid in the feed had forced most of the zirconium through to the sorption cycle effluent together with a large part of the ruthenium (see Tables 3 and 10). Consequently the wash cycle contained negligible amounts of zirconium but appreciable quantities of ruthenium. In Runs 21-25 of Table 10, the wash cycle was increased from two to three resin bed volumes. Comparison of the gamma spectra from these wash cycle solutions with those for Runs 16-19 showed that additional Ru-106 was eluted. However, spectrophotometric analysis of the third bed volume revealed that a further 1 per cent. of the beryllium was also eluted. Therefore a two bed volume wash of 0.05M oxalic acid was adopted for all subsequent separations.

4.5 Elution Cycle

The aim of elution is the selective concentration of the beryllium in the effluent, with a minimum of contamination from fission products. This can be achieved with an eluent which contains an anion which selectively complexes the beryllium. If the cation concentration remains low, for example with a weak acid, fission product movement down the column is kept to a minimum.

Investigation of the sorption isotherms of beryllium (Fardy 1965) has shown that hydrofluoric acid readily complexes beryllium. This acid, and carboxylic acids, were tested on various size columns saturated with beryllium. The results of this study are contained in Table 12. The choice of suitable carboxylic acids was based on the work of Fardy (1962) and de Bruin, Kairiatis, and Temple (1962).

The carboxylic acids did not elute the beryllium rapidly until the pH of the eluents was raised to 4.0. However hydrofluoric acid elutes the beryllium as rapidly as these acids and has the added advantage of possessing low cation concentration. Although 0.25M triammonium citrate did elute the beryllium in three bed volumes this eluent was discarded. This decision was based on the relative concentration of the ammonium ion and hydrogen ions in the respective eluent and the higher affinity of the ammonium ion for the cation exchange resin. Furthermore, the radiation stability of inorganic eluents makes them an obvious choice over organic compounds.

The optimum acid concentration used in the separation studies was determined on the chromatographic column packed with Zeo-Karb 225, 50 - 100 mesh, cation-exchange resin. Elution data were obtained for various concentrations of hydrofluoric acid but with the resin loaded to its practical capacity with beryllium. The unusual hydrofluoric acid concentration figures, shown in Table 13, resulted from the use of 5 ml and 10 ml Nalgene pipettes for handling the concentrated hydrofluoric acid. Standardisation of the solutions prepared from these pipettes yielded the figures shown. Since the cation concentration and the number of bed volumes of the eluent must be kept to a minimum, 0.46M hydrofluoric acid was selected for use in all separation studies.

4.6 Regeneration Cycle

The recommendation of Blanco and his co-workers (1956) regarding the use of 6M HNO₃ in the regeneration cycle was tested and adopted. Subsequent column testing showed that a five bed volume wash with acid of this concentration removed most of the activity from the column.

4.7 Change in Column Geometry

After it had been shown that a gamma decontamination factor of 2×10^3 could be obtained, using a chromatographic column and optimum conditions, an investigation was made of the change in fission product separation produced by a change in column dimensions. A summary of this investigation, which was made under the same optimum conditions, is contained in Table 14. Increasing the column diameter to 1.30 cm but maintaining a constant bed volume and flow rate (1 ml/min.cm²) resulted in a much faster process time. However, the gamma D.F. was reduced to 4×10^2 . Furthermore, experiments revealed that the breakthrough capacity of this shorter column had decreased by 10 per cent. Gamma spectrometer analysis showed the cause of the poor fission product separation to be the elution of Cs-137 with the beryllium.

Maintaining a constant diameter of 1.30 cm for the column but increasing the length from 11.0 cm to 48.0 cm restored the gamma D.F. figure to that obtained previously. Thus the length of the column is important in this type of separation but a change in the diameter exerts little influence.

Further studies with the smaller column (11.0 cm x 1.3 cm) showed that the inclusion of a small auxiliary zirconium phosphate column during the sorption cycle prevented Cs-137 contamination of the beryllium in the elution cycle. However 20 per cent. of this auxiliary column was dissolved through its interaction with the oxalic acid. This reaction did not impede the separation by blocking the column but instead aided decontamination by sweeping more Zr-95 and Ru-106 from the column. Thus a gamma D.F. of 3×10^3 was obtained for this run. Dissolution of the

auxiliary column may be overcome by passing the beryllium feed at pH 1.0 through the zirconium phosphate column, adding the oxalic acid, and then proceeding with the separation in the normal manner.

4.8 Possible Feed Pretreatment

One of the standard practices for removing ruthenium and zirconium from reactor fuel processing solutions is by scavenging precipitation. Blanco and co-workers (1956) included this step in their process for aluminium recovery by ion exchange and were able to obtain a gamma decontamination factor of 2×10^3 . The addition of oxalic acid to a beryllium feed enabled a similar decontamination to be obtained for beryllium recovery by a pure ion-exchange process (see Section 4.3.3). However, ruthenium and zirconium were the contaminants that limited this decontamination to a factor of 2×10^3 .

A similar scavenging process was tested with the ion-exchange separation in the absence and presence of oxalic acid (Table 15). The results clearly showed that the inclusion of a scavenging pretreatment did nothing to enhance the separation.

5. OUTLINE OF PROPOSED PROCESS

On the basis of the results reported above, a separation process has been developed for beryllium in waste solutions contaminated with fission products.

The ion-exchange process involves the sorption of beryllium and fission products on a cation-exchange resin in the presence of oxalic acid, and, after a suitable wash cycle, selective elution of the beryllium with hydrofluoric acid. The steps in the process are set out below, and a flow sheet is given in Figure 8.

1. The beryllium concentration in the feed is diluted to 0.1M and the solution made 0.1M in oxalic acid. The pH is adjusted to 1.
2. The solution is passed through a column (48.0 cm x 0.60 cm) of Zeo-Karb 225, 50 - 100 mesh, 8% D.V.B., hydrogen-form resin until the resin is saturated to breakthrough loading with respect to beryllium.
3. The column is washed with two bed volumes of 0.05M oxalic acid.
4. The beryllium is selectively eluted with five bed volumes of 0.46M hydrofluoric acid.
5. Regeneration of the column is accomplished with five bed volumes of 6M HNO_3 .
6. After being rinsed with demineralised water the column is ready for re-use.

Typical gamma spectra obtained at various stages of the above process are shown in Figures 9 - 12.

6. CONCLUSION

The use of cation-exchange resin to separate and decontaminate beryllium from an aqueous nitrate solution containing fission products has been demonstrated on the laboratory scale. A 97 per cent. recovery of beryllium with gross beta and gamma decontamination factors of 10^4 and 2×10^3 respectively was obtained. The process is flexible and should be applicable to both sulphate and chloride media.

7. REFERENCES

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TABLE 1

SORPTION OF FISSION PRODUCTS BY CATION-EXCHANGE
RESINS IN THE BERYLLIUM FORM AS A FUNCTION OF
BERYLLIUM CONCENTRATION (pH = 1.0)

Be Conc'n	DISTRIBUTION COEFFICIENT									
	Cs-137		Sr-85		Ce-144		Zr-95		Ru-106	
	A	B	A	B	A	B	A	B	A	B
0.1M	16.5	23.0	116.6	26.2	2131.5	278.4	25.4	422.8	19.2	22.0
0.2M	10.0	19.5	57.3	14.1	713.9	91.8	33.4	258.0	15.8	14.6
0.5M	6.5	9.5	15.7	4.2	114.9	15.8	5.4	277.0	3.6	7.3
1.0M	4.0	7.0	4.5	2.6	23.5	5.7	8.7	170.5	0.5	2.5
2.0M	0.7	3.5	1.0	1	3.6	<1.5				

A : Zeo-Karb 225, 20 - 50 mesh, Be form.

B : Zeo-Karb 215, 20 - 50 mesh, Be form.

TABLE 2

SORPTION OF FISSION PRODUCTS BY THE HYDROGEN
FORM OF THE CATION EXCHANGER, ZEO-KARB 225, FROM
0.1M BERYLLIUM SOLUTION, AS A FUNCTION OF RESIN CROSS-
LINKAGE

Fission Product	DISTRIBUTION COEFFICIENT			
	High Porosity 4-5% DVB	Medium Porosity 8% DVB	Low Porosity 12% DVB	Resin in Be ⁺² Form 8% DVB
Cs-137	17.1	35.2	54.6	16.5
Sr-85	49.5	178.9	282.8	117.0
Ce-144	267.1	6104.2	5530	2131.5
Ru-106	30.8	26.9	29.2	32.5
Zr-95	21.3	21.5	29.3	20.8

TABLE 3
SUMMARY OF SEPARATIONS
THE CHANGE IN BETA AND GAMMA D.F.* WITH INCREASING
BERYLLIUM CONCENTRATION IN THE FEED

Run No.	Feed and Pretreatment	Resin Column	Column Saturation	Wash Cycle	Elution Cycle	% Be Recovered	Overall Beta and Gamma D.F.*
16-19	0.1M Be, 0.1M Oxalic Acid, pH 1.0	Zeo-Karb 225 50 - 100 mesh, 8% DVB H ⁺ form Column Dimensions 48.0 cm x 0.62 cm	95%	0.05M Oxalic Acid	0.46M HF	96	$\beta = 10^4$ $\gamma = 10^3$
26-27	0.2M Be, 0.1M Oxalic Acid, pH 1.0	As above	96%	As above	As above	98	$\beta = 10^4$ $\gamma = 9 \times 10^2$
35-36	0.5M Be, 0.1M Oxalic Acid, pH 1.0	As above	92%	As above	As above	97	$\gamma = 1.2 \times 10^3$

* D.F. is the decontamination factor and is defined as:

Decontamination Factor for Contaminant "i"

$$= \frac{\text{Concentration of i in feed/concentration of valued constituent in feed}}{\text{Concentration of i in product/concentration of valued constituent in product}}$$

TABLE 4
SORPTION OF FISSION PRODUCTS BY CATION-EXCHANGE
RESINS IN THE BERYLLIUM FORM FROM 0.2M BERYLLIUM
SOLUTIONS AS A FUNCTION OF pH

pH	DISTRIBUTION COEFFICIENT									
	Cs-137		Sr-85		Ce-144		Zr-95		Ru-106	
	A	B	A	B	A	B	A	B	A	B
0.5	14.0	15.2	37.0	9.1	34.0	40.2	20.8	665.6	7.6	9.8
1.0	19.3	20.2	55.4	13.7	754.1	88.1	27.0	514.8	9.1	12.9
1.5	18.5	16.9								
2.0	18.7	19.9	52.9	14.1	937.5	114.6	23.2	392.8	16.4	24.0
2.5	18.7	19.2								
3.0	19.0	20.2	37.4	14.1	489.8	84.8	32.2	186.0	58.5	66.6

A Zeo-Karb 225, 20 - 50 mesh, Be form.

B Zeo-Karb 215, 20 - 50 mesh, Be form.

TABLE 5

**SORPTION OF Ru-106 BY CATION-EXCHANGE RESINS IN
THE BERYLLIUM FORM IN THE PRESENCE AND ABSENCE
OF THIOUREA**

Be Conc'n	pH	Complex Agent	Resin (20-50 Mesh)	Distribution Coefficient
0.2M	1.0	Nil	Zeo-Karb 225	20.0
0.2M	1.0	0.2% thiourea	Zeo-Karb 225	231.6
0.2M	1.0	0.2% thiourea	Zeo-Karb 215	987.0
0.1M	3.0	Nil	Zeo-Karb 225	82.0
0.1M	3.0	Nil	Zeo-Karb 215	91.5
0.1M	3.0	0.2% thiourea	Zeo-Karb 225	426.3
0.1M	3.0	0.2% thiourea	Zeo-Karb 215	810.8

TABLE 6

**FISSION PRODUCT SORPTION FROM 0.1M BERYLLIUM
SOLUTION BY THE CATION EXCHANGER
ZEO-KARB 225 20-50 MESH, IN THE PRESENCE OF THIOUREA**

Radionuclide	Distribution Coefficient in 0.2% Thiourea	Distribution Coefficient Without Thiourea
Cs-137	25.2	26.5
Sr-85	84.9	116.6
Ce-144	1957.5	2131.5
Zr-95	76.2	25.4
Ru-106	272.1	19.2

TABLE 7
SUMMARY OF SEPARATIONS
COLUMN TESTING OF THIOUREA

Run No.	Feed and Pretreatment	Resin and Column	Column Saturation	Wash Cycle	Elution Cycle	% Be Recovered	Overall Beta and Gamma D.F.
12-13	0.5M Be Addition of Thiourea 0.2%. Heat to 90°C for 1 hour. Dilution to 0.1M Be, pH 1.0	Zeo-Karb 215, 20-50 mesh, H ⁺ form, Column Dimensions 3.0 cm x 0.76 cm	100%				
		Zirconium Phosphate, 60-120 mesh, H ⁺ form, Column Dimensions 2.0 cm x 0.76 cm	Nil				
		Zeo-Karb 225, 50-100 mesh, 8% DVB H ⁺ form	87%	0.02M	0.46M	92	$\beta = 4 \times 10^2$ $\gamma = 5 \times 10^1$
14-15	0.5M Be Addition of Thiourea 0.2%. Heat to 90°C for 1 hour. Dilution to 0.1M Be, pH 1.0	Silica Gel, 40-80 mesh, Column Dimensions 10.50 cm x 0.76 cm As for Run Nos. 12-13	87%	0.02M Oxalic Acid	0.46M HF	95	$\beta = 7 \times 10^2$ $\gamma = 2 \times 10^2$
16-19	0.1M Be, 0.1M Oxalic Acid, pH 1.0	Zeo-Karb 225, 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 48.0 cm x 0.62 cm	95%	0.05M Oxalic Acid	0.46M HF	96	$\beta = 10^4$ $\gamma = 10^3$

TABLE 8

SORPTION OF BERYLLIUM AND ZIRCONIUM BY CATION
EXCHANGE-RESINS IN THE PRESENCE AND ABSENCE OF
0.1M OXALIC ACID (pH = 1.0)

Solution	Resin	Distribution Coefficients
Be-7, 0.1M HNO ₃	Zeo-Karb 225 H ⁺ form	1382
Be-7, 0.1M Oxalic Acid, pH = 1.0	"	1139
Zr-95, 0.1M Be, pH = 1.0	"	20.8
Zr-95, 0.1M Be, 0.1M Oxalic Acid	"	< 1.0
Zr-95, 10 ⁻⁴ M Zr, 0.1M Be, 0.1M Oxalic Acid	"	< 1.0
Zr-95, 10 ⁻⁴ M Zr, 0.1M Be, 0.1M Oxalic Acid	Zeo-Karb 215 H ⁺ form	< 1.0

TABLE 9

SORPTION OF FISSION PRODUCTS BY THE CATION
EXCHANGER ZEO-KARB 225, HYDROGEN FORM, FROM
0.1M BERYLLIUM CONTAINING 0.1M OXALIC ACID (pH=1.0)

Solutions	Distribution Coefficient (Oxalic Acid)	Distribution Coefficient (Nitric Acid)
Cs-137	31.6	27.5
Sr-85	132.8	120.6
Ce-144	354.6	2131.5
Ru-106	< 1	19.2

TABLE 10

SUMMARY OF SEPARATIONS

COLUMN TESTING OF OXALIC ACID ADDITION TO THE FEED

Run No.	Feed and Pretreatment	Resin Column	Column Saturation	Wash Cycle	Elution Cycle	% Be Recovered	Overall Beta and Gamma D.F.
16-19	0.1M Be, 0.1M Oxalic Acid, pH = 1.0	Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 48.0 cm x 0.62 cm	95%	0.05M Oxalic Acid (Two Bed Vols)	0.46M HF	96	$\beta = 10^4$ $\gamma = 10^3$
21-25	0.1M Be, 0.1M Oxalic Acid, pH = 1.0	Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 48.0 cm x 0.62 cm	96%	0.05M Oxalic Acid (Three Bed Vols)	0.46M HF	97	$\gamma = 2 \times 10^3$

TABLE 11

**BERYLLIUM LOSS BY VARIOUS OXALIC ACID CONCENTRATIONS
DURING WASH CYCLE**

RESIN: ZEO-KARB 225, 50-100 MESH, 8% DVB

Oxalic Acid Concentration	% Beryllium Eluted	Column Saturation
0.02M	< 0.5	Breakthrough loading
0.05M	1.0	„ „
0.10M	5.1	„ „
0.10M	5.5	Practical loading
0.05M	1.1	Breakthrough loading
0.05M	2.1	Practical loading

TABLE 12

THE ELUTION OF BERYLLIUM (%) BY VARIOUS ELUENTS FROM
THE CATION EXCHANGER ZEO-KARB 225 20-50 MESH 8% DVB

Eluent	Bed Volume Number					
	3	5	7	9	12	15
0.4M HNO ₃ (Bed Vol. = 18 ml)	43.0	55.9		70.0		81.5
0.4M HNO ₃ (Bed Vol. = 32.5 ml)	43.0	56.6		70.0	76.5	80.9
0.5M HNO ₃	47.2		78.5			99.2
0.25M Oxalic Acid (pH 4.0)	64.4	101				
0.46M Oxalic Acid	33.4	42.1	48.5	53.1	57.4	
0.25M Malonic Acid (pH 4.0)	36.0	70.0		102.0		
0.46M Malonic Acid (pH 4.0)	60.6	100.0				
0.25M Salicylic Acid - 0.25M NaOH		68.1		97.2		
0.25M Sulphosalicylic Acid (pH 4.0)	69.6	102.0				
0.46M Sulphosalicylic Acid	46.5	61.1		73.9	81.7	
0.25M Triammonium Citrate	98.7					
0.24M Hydrofluoric Acid	30.6	57.6		97.2		
0.46M Hydrofluoric Acid	72.7	104.0				
0.25M Ammonium Fluoride	31.7	60.6		98.5		

TABLE 13

BERYLLIUM (%) ELUTION FROM THE CATION EXCHANGER
ZEO-KARB 225 50-100 MESH BY VARIOUS CONCENTRATIONS OF
HYDROFLUORIC ACID

Eluent	Bed Volume Number						
	1	2	3	4	5	7	9
0.236M HF	3.1		26.1		49.5	73.1	98.2
0.46M HF	12.3		59.4	90	100		
0.46M HF	12.2		66.5	92	100		
0.67M HF	21.5	57.0	91.6	100			
0.67M HF	49.5	59.0	92.0	100			

TABLE 14

SUMMARY OF SEPARATIONS

THE CHANGE IN D.F. WITH CHANGING COLUMN DIMENSIONS

Run No.	Feed and Pretreatment	Resin Column	Column Saturation	Wash Cycle	Elution Cycle	% Be Recovered	Overall Beta and Gamma D.F.
16-19	0.1M Be, 0.1M Oxalic Acid, pH 1.0	Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 48.0 cm x 0.62 cm	95%	0.05M Oxalic Acid	0.46M HF	96	$\beta = 10^4$ $\gamma = 10^3$
30-32	As above	Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 11.0 cm x 1.29 cm	96%	0.05M Oxalic Acid	0.46M HF	97	$\gamma = 4 \times 10^2$
32-33	As above	Zirconium Phosphate, 60-120 mesh, H ⁺ form, Column Dimensions 3.25 cm x 0.76 cm Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 11.0 cm x 1.29 cm	96%	0.05M Oxalic Acid	0.46M HF	97	$\gamma = 3 \times 10^3$
34	As above	Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 48.0 cm x 1.29 cm	96%	0.05M Oxalic Acid	0.46M H ⁺	97	$\gamma = 10^3$

TABLE 15

SUMMARY OF SEPARATIONS

THE USE OF SCAVENGING PRECIPITATION

Run No.	Feed and Pretreatment	Resin Column	Column Saturation	Wash Cycle	Elution Cycle	% Be Recovered	Overall Beta and Gamma D.F.
37-38	0.5M Be Feed Scavenging Pptn. 0.01% Fe(III) 0.01% Mn(IV) Centrifuged dilution to 0.1M Be	Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 47.0 cm x 0.62 cm	85%	0.02M Oxalic Acid	0.46M HF	99	$\beta = 10^3$ $\gamma = 2 \times 10^2$
39-41	0.5M Be Feed Scavenging Pptn. 0.05% Fe(III) 0.05% Mn(IV) Centrifuged, Supernatant dilution to 0.1M Be, pH 1.0	Zirconium Phosphate, 60-120 mesh, H ⁺ form Column Dimensions 37.3 cm x 0.76 cm Zeo-Karb 225 50-100 mesh, 8% DVB H ⁺ form Column Dimensions 47.0 cm x 0.62 cm	Nil 85%	Nil 0.02M Oxalic Acid	Nil 0.46M HF	 93	 $\beta = 4 \times 10^3$ $\gamma = 10^3$
42-43	0.5M Be Feed Scavenging Pptn. 0.04% Fe(III) 0.05% Mn(IV) Centrifuged, Supernatant dilution to 0.1M Be, 0.1M Oxalic Acid, pH 1.0	Zeo-Karb 225 50-100 mesh, 7-9% DVB, H ⁺ form Column Dimensions 48.0 cm x 0.62 cm	96%	0.05M Oxalic Acid	0.46M HF	97	$\gamma = 10^3$
16-19	0.1M Be, 0.1M Oxalic Acid, pH 1.0	Zeo-Karb 225 50-100 mesh, 7-9%DVB H ⁺ form Column Dimensions 48.0 cm x 0.62 cm	95%	0.05M Oxalic Acid	0.46M HF	96	$\beta = 10^4$ $\gamma = 10^3$

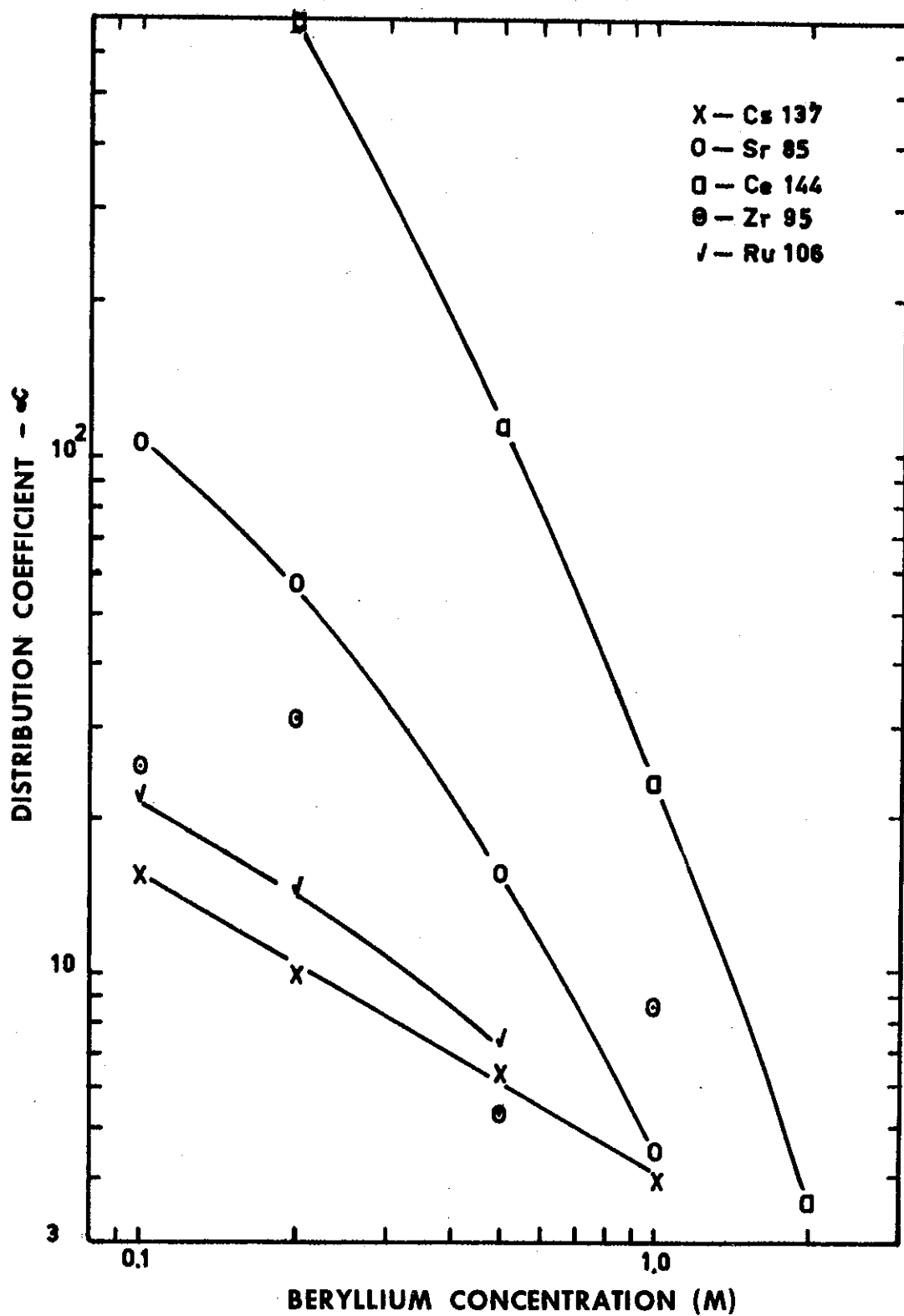


FIGURE 1 SORPTION OF FISSION PRODUCTS BY THE BERYLLIUM FORM OF THE CATION EXCHANGER ZEO-KARB 225 AS A FUNCTION OF BERYLLIUM CONCENTRATION

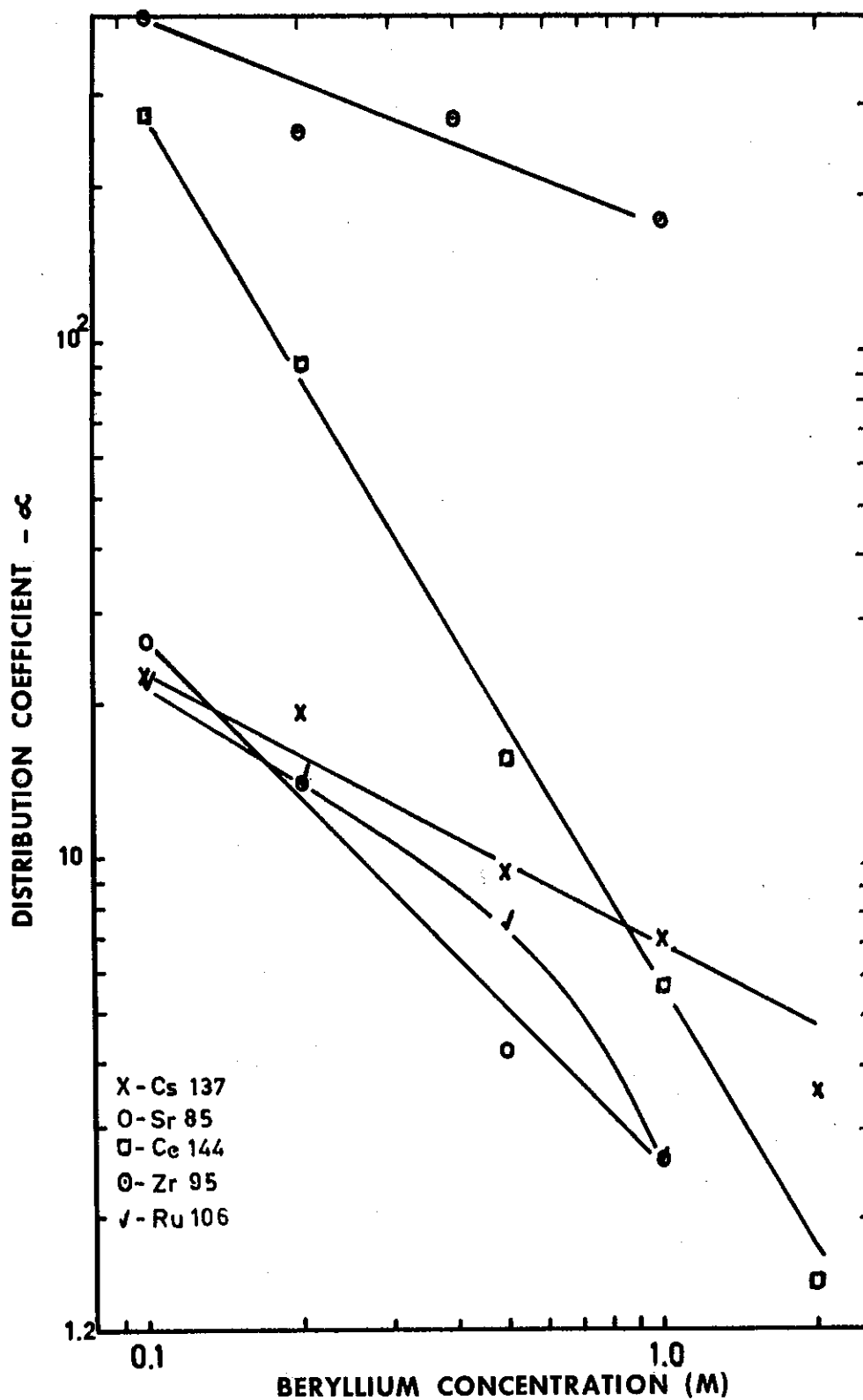


FIGURE 2 SORPTION OF FISSION PRODUCTS BY THE BERYLLIUM FORM OF THE CATION EXCHANGER ZEO-KARB 215 AS A FUNCTION OF BERYLLIUM CONCENTRATION

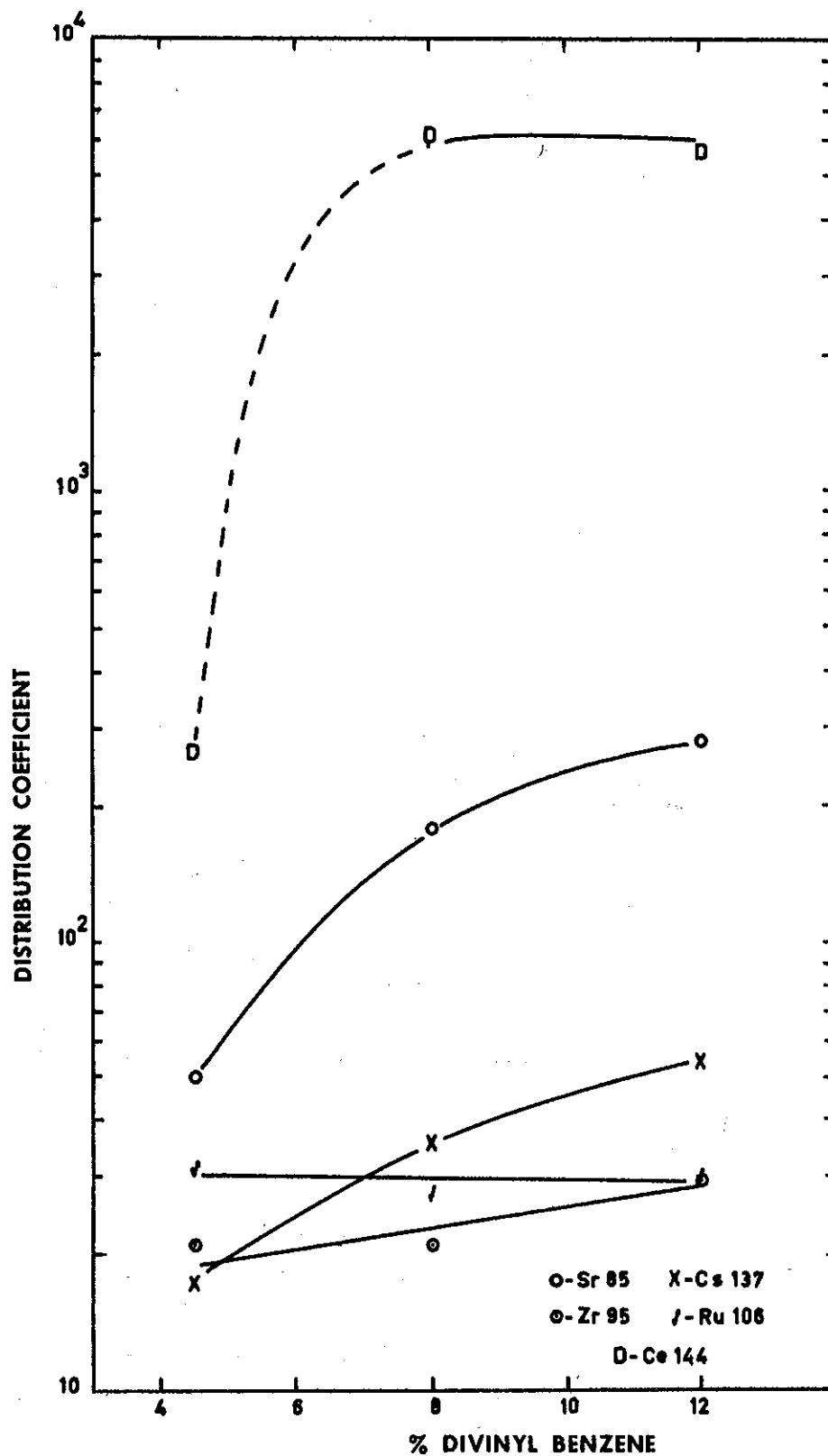


FIGURE 3 SORPTION OF FISSION PRODUCTS BY THE HYDROGEN FORM OF THE CATION EXCHANGER ZEO-KARB 225 FROM 0.1M BERYLLIUM SOLUTIONS AS A FUNCTION OF CROSS-LINKAGE

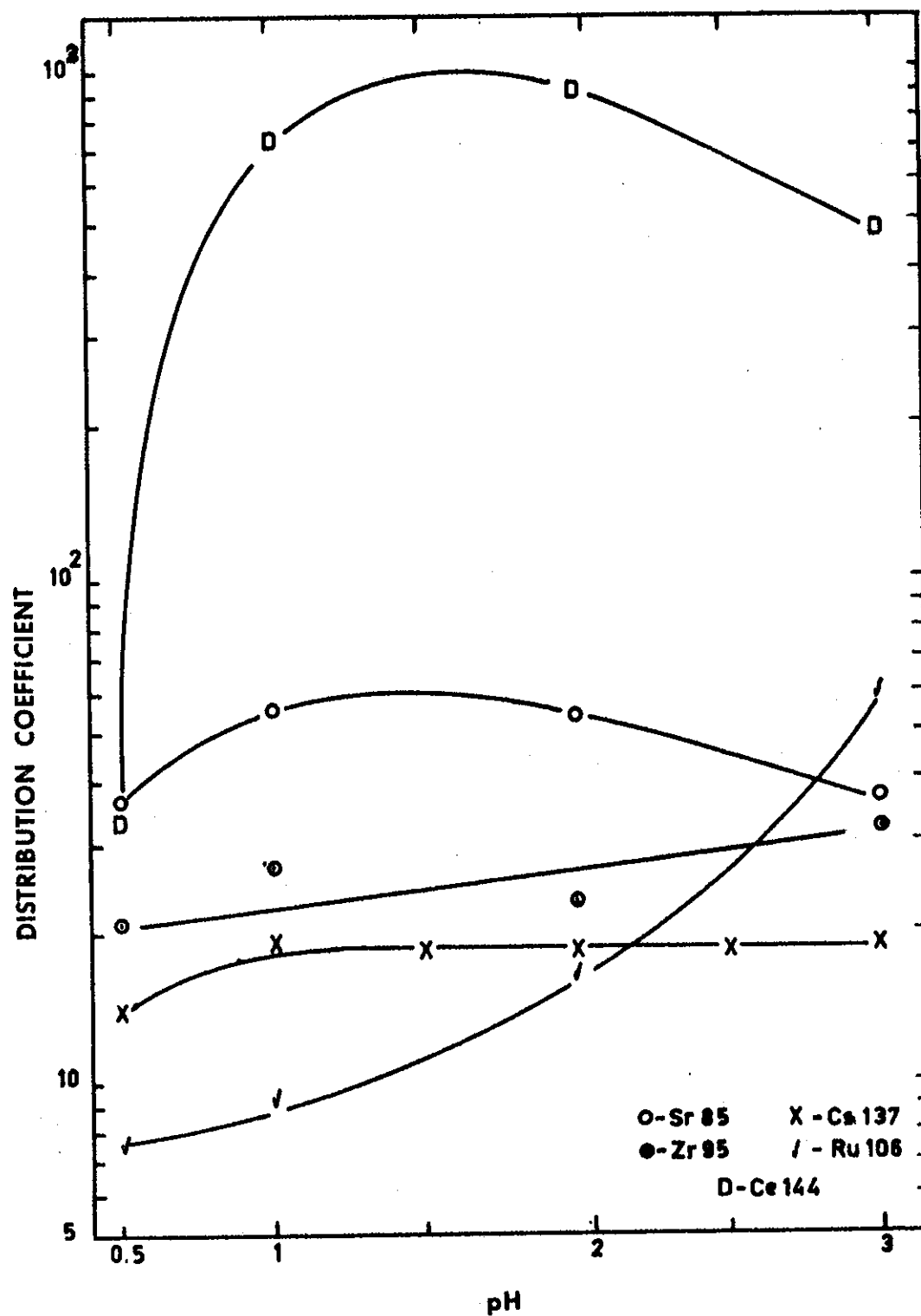


FIGURE 4 SORPTION OF FISSION PRODUCTS BY THE
CATION EXCHANGER ZEO-KARB 225

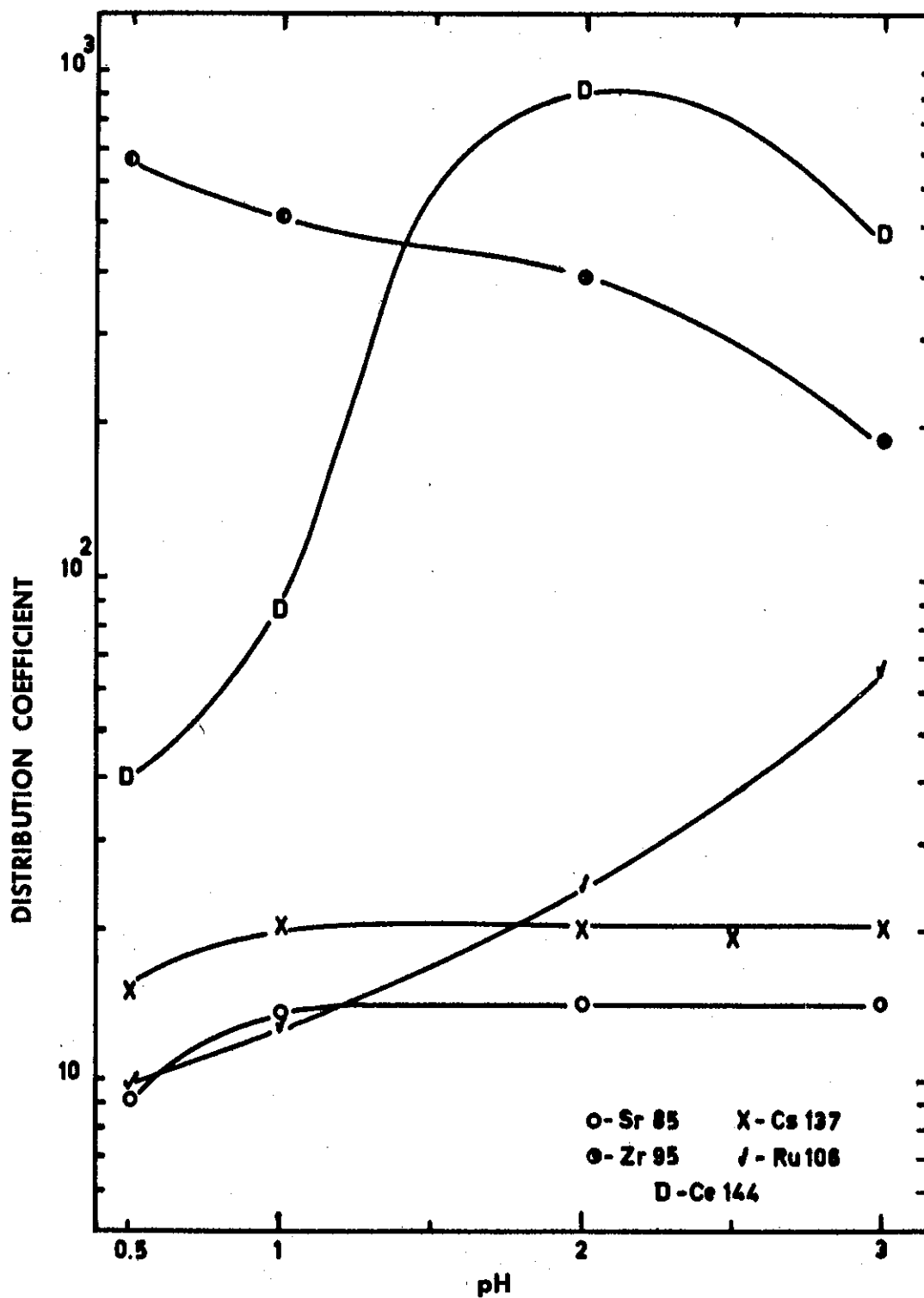


FIGURE 5 SORPTION OF FISSION PRODUCTS BY THE CATION EXCHANGER ZEO-KARB 215 IN THE BERYLLIUM FORM FROM 0.2M BERYLLIUM SOLUTION AS A FUNCTION OF pH

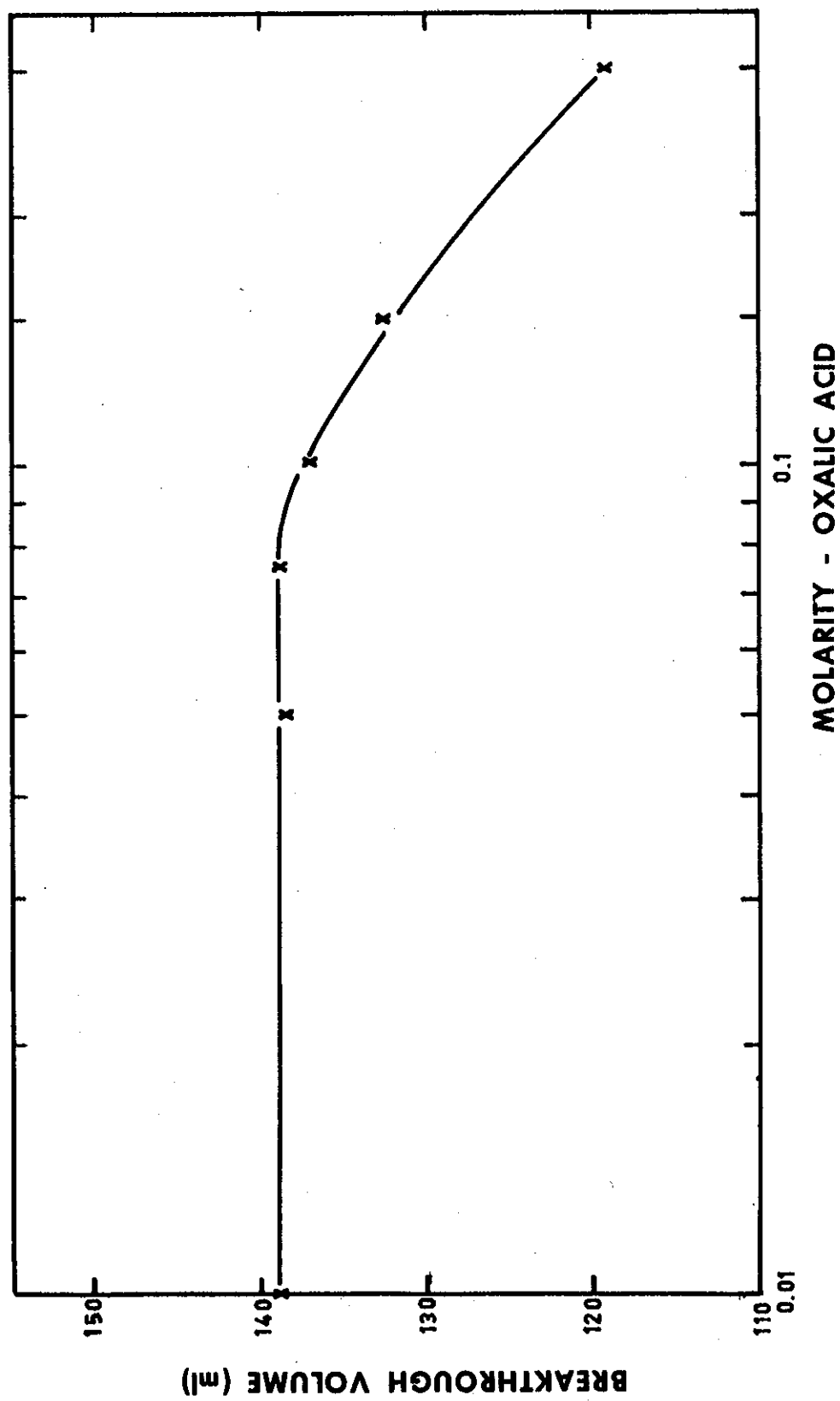
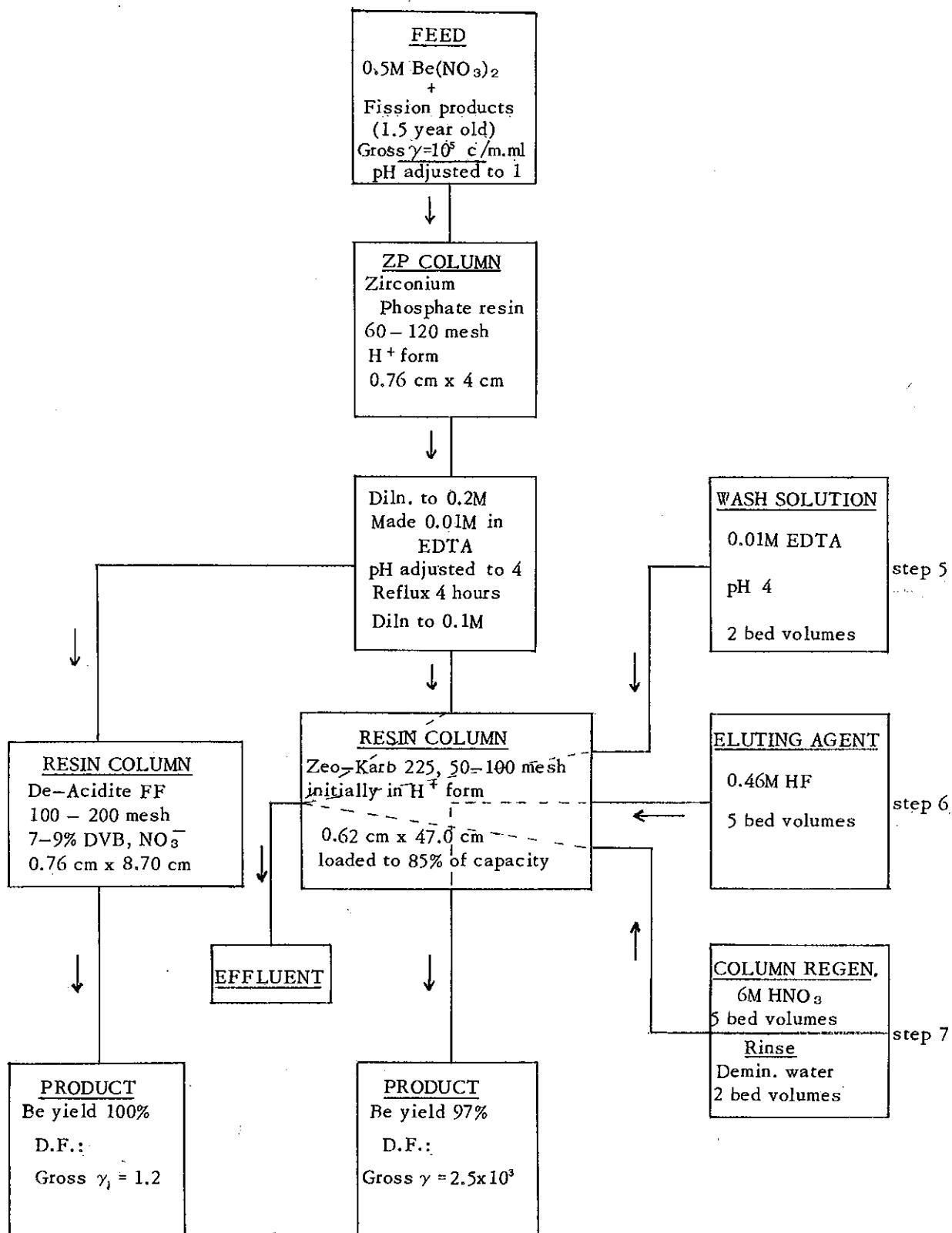


FIGURE 6 BREAKTHROUGH VOLUME FOR 0.1M BERYLLIUM FEED AT pH 1.0 CONTAINING VARIOUS CONCENTRATIONS OF OXALIC ACID ON ZEO-KARB 225, H^+ FORM, 50-100 MESH



**FIGURE 7 FLOWSHEET FOR BERYLLIUM DECONTAMINATION
USING EDTA**

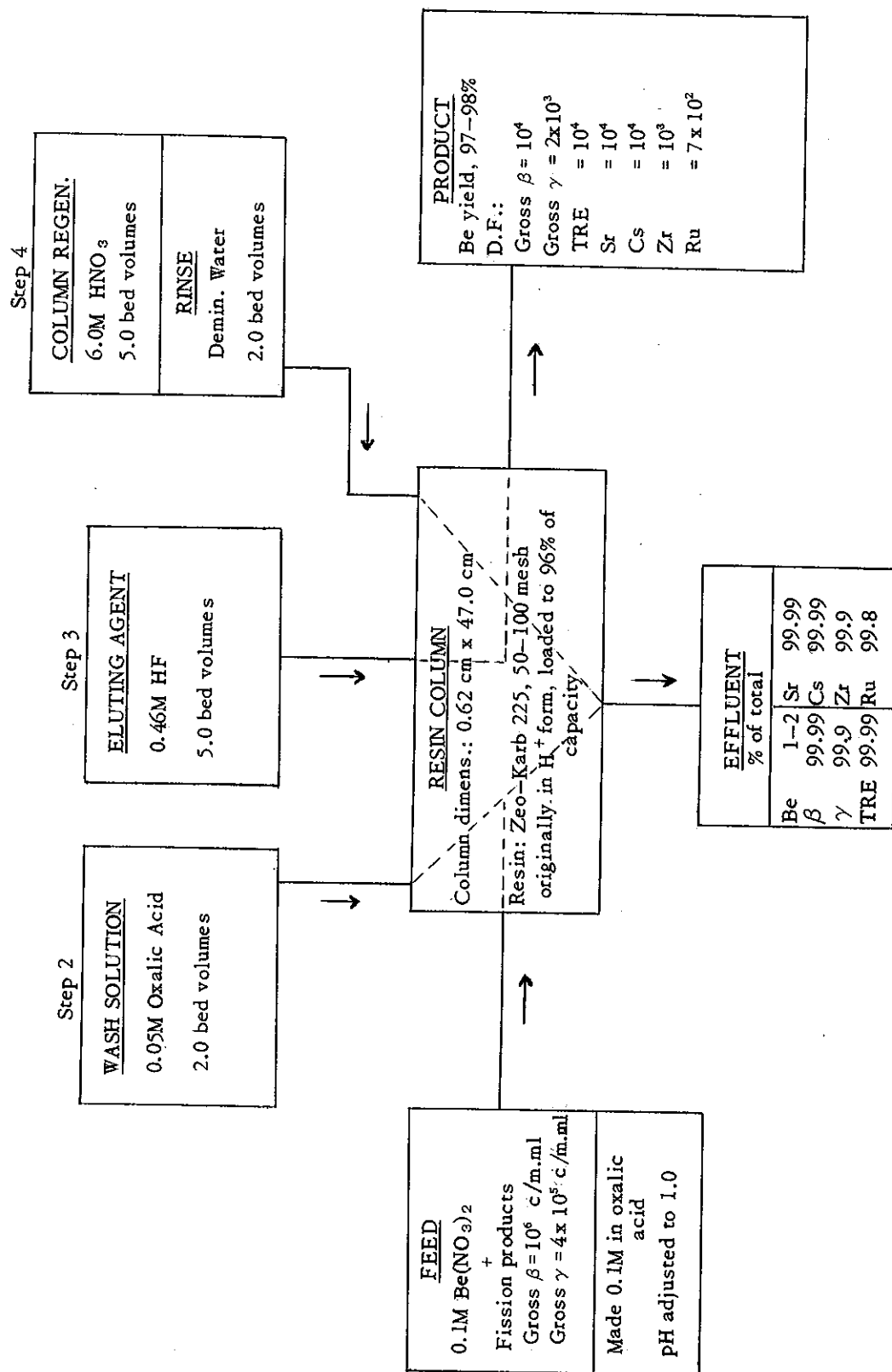


FIGURE 8 FLOWSHEET FOR BERYLLIUM DECONTAMINATION
USING OXALIC ACID

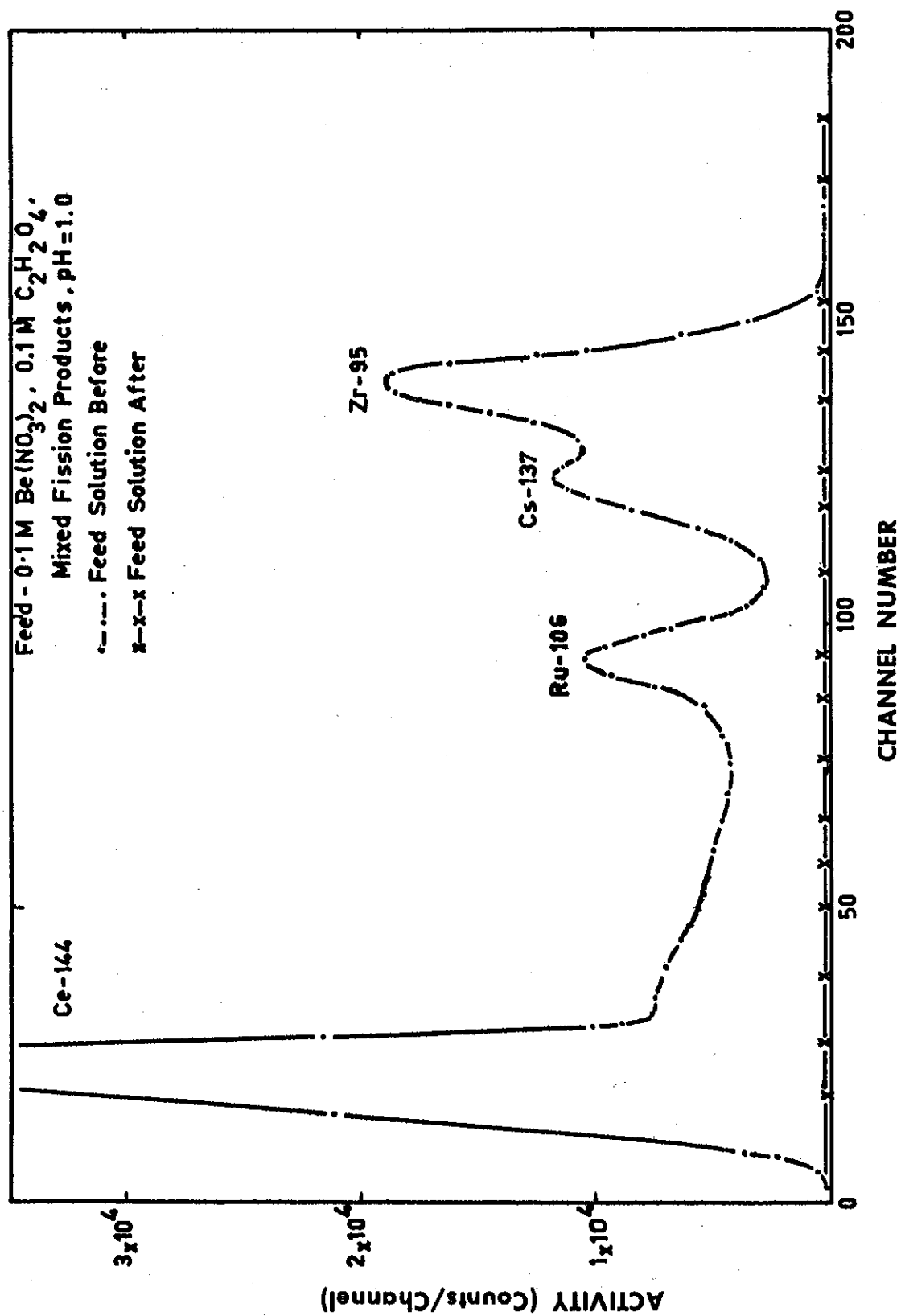


FIGURE 9 GAMMA SPECTRA OF 0.1M BERYLLIUM SOLUTION BEFORE
AND AFTER ION-EXCHANGE TREATMENT

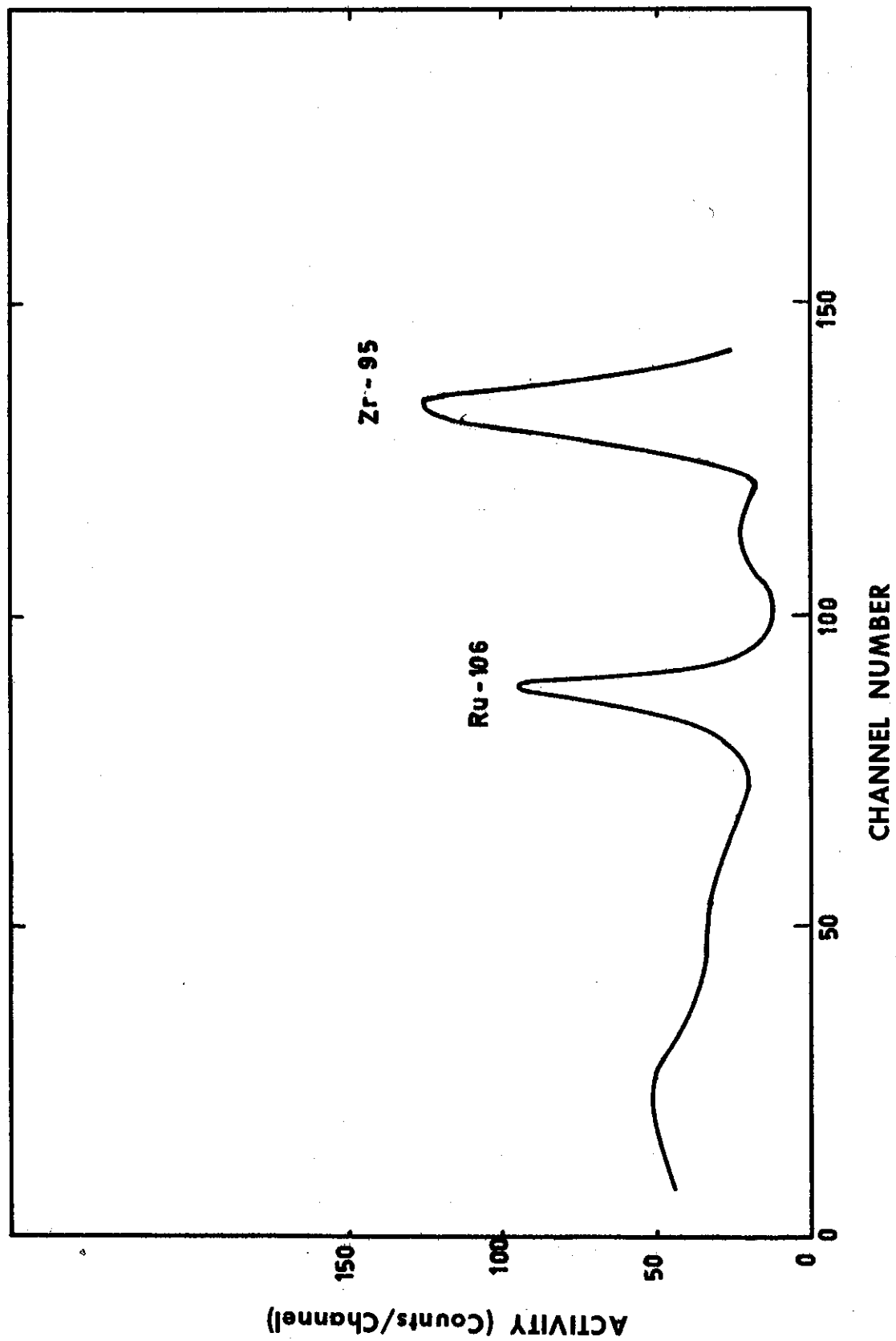


FIGURE 10 GAMMA SPECTRUM OF ELUTION CYCLE EFFLUENT

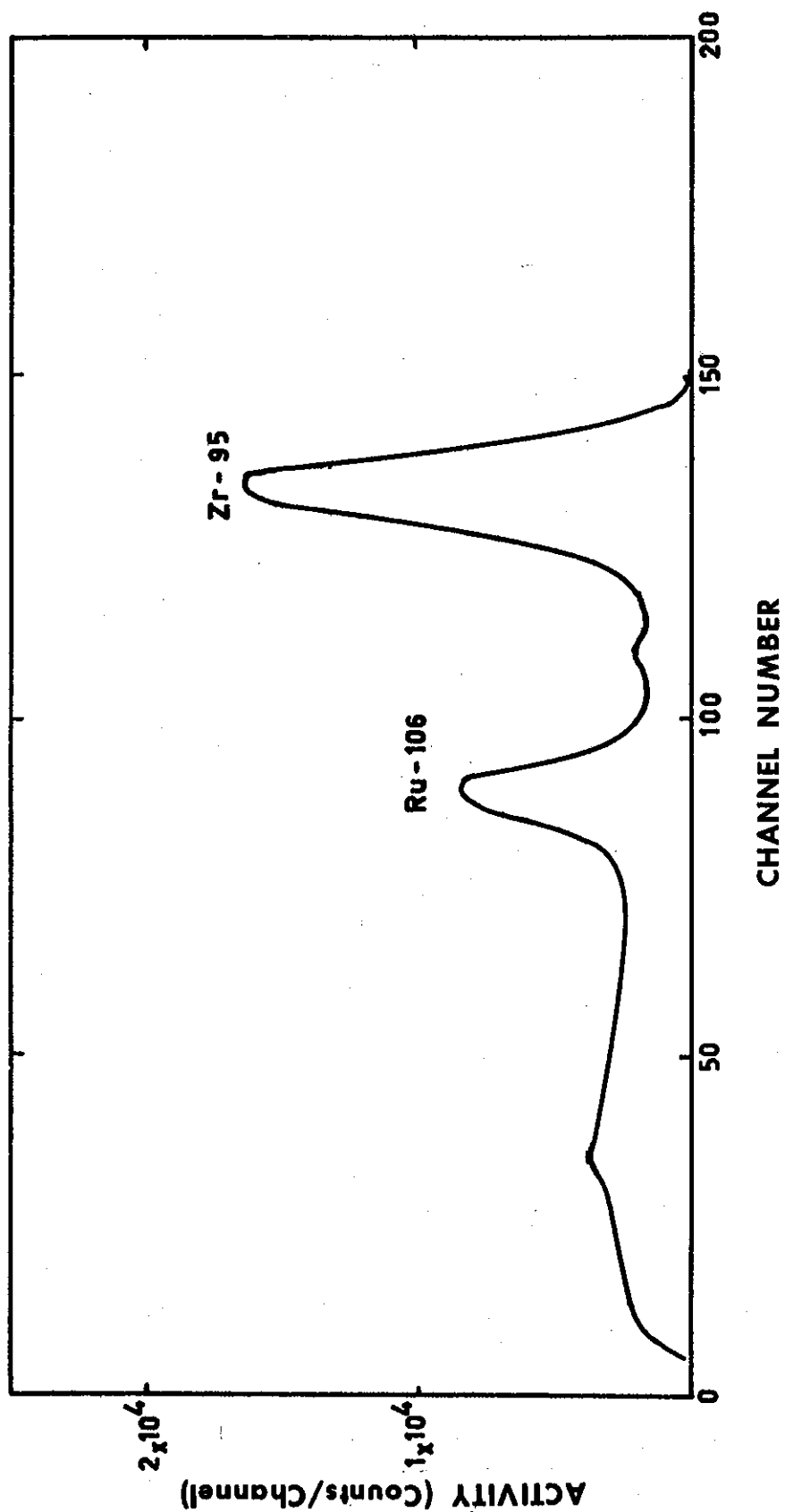


FIGURE 11 GAMMA SPECTRUM OF SORPTION CYCLE EFFLUENT --
MAJORITY OF RU-106 AND Zr-95 HAS PASSED TO THE EFFLUENT

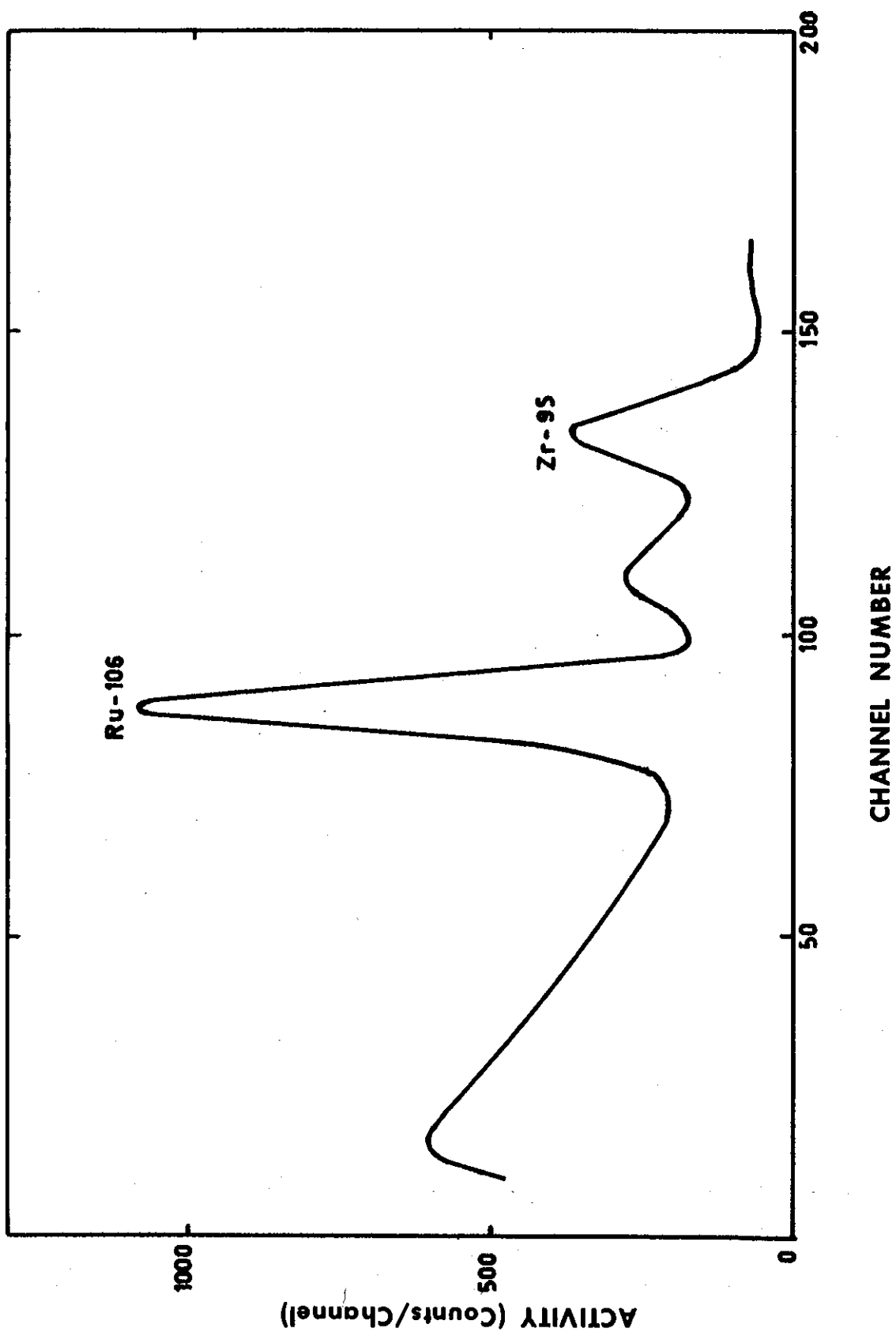


FIGURE 12 GAMMA SPECTRUM OF 0.05M OXALIC ACID WASH CYCLE EFFLUENT --
(TWO BED VOLUMES). MAINLY RU-106 WITH A SMALL AMOUNT OF Zr-95