1956

A procedure for the estimation of total radioactivity in water

Kenneth Vergil Marsh
University of the Pacific

A PROCEDURE FOR THE ESTIMATION OF TOTAL RADIOACTIVITY IN WATER

A Thesis

Presented to

the Faculty of the Department of Chemistry

College of the Pacific

In Partial Fulfillment

of the Requirements for the Degree

Master of Science

---

by

Kenneth Vergil Marsh

June 1956

ACKNOWLEDGMENT

The author wishes to express his appreciation to Dr. William H. Wadman whose patience, tolerance, consolation, and technical assistance were of invaluable aid throughout the entire course of this research.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

CHAPTER I

INTRODUCTION AND STATEMENT OF THE PROBLEM

One of the essential duties of the State of California Department of Civil Defense in times of nuclear weapon attack will be radiological monitoring. The workers in the field will operate from a mobile laboratory truck, equipped with radiation counters and meters, but containing only the simplest of chemical equipment. One of their main problems will be the determination of the total amount of radioactive material, especially fallout of fission products, in a water sample. As yet no method of analysis both simple and rapid enough for application to mobile operation has been developed.

It was the purpose of this research to develop and refine a simple, rapid, quantitative procedure for the detection and estimation of the total amount of radioactive material contained in a water sample, using only readily available and preferably inexpensive apparatus. However, no attempt has been made to either qualitatively or quantitatively determine any of the possible individual constituents.

CHAPTER II

REVIEW OF THE LITERATURE

Much has been written in regard to analysis of fission products, their separation, relative proportions, activities, and occurrence, and several investigators have applied ion exchange principles to their own particular problems. However, only a brief summary of the work done on those problems most closely related to the one at hand will be given here.

I. ANALYSIS AND SEPARATION OF FISSION PRODUCTS-- ION EXCHANGE

Pioneer work on the absorption of metal ions on an exchange resin as a means of separating fission-produced isotopes was done by Tompkins, Khym, and Cohn (J. Am. Chem. Soc. 1947) who found the following relative strengths of absorption on a cation resin: Th>La>Ce>Rare Earths > Y>Ba>Cs>Sr>K>NH_4>H. They also discuss the advantages of column over batch operations on both practical and chemical principles. A table is given showing the major fission products coexisting in uranium about one month from the end of a several month exposure in a chain reacting pile. More work on this problem has been done by Ketelle and

Boyd (J. Am. Chem. Soc. 1951). The method has undergone considerable refinements, and more recent papers (Brown and Rieman, 1952; Wheelwright and Spedding, 1953: Freiling and Bunney, 1954) have elaborated on techniques. Finally, Downing, Wheatland and Eden (J. Inst. Water Engrs., 1953) showed that ion exchange removed over 97 per cent of radioactive contaminants--a higher percentage than any other process.

II. ANALYSIS AND SEPARATION OF FISSION PRODUCTS BY METHODS OTHER THAN ION EXCHANGE

A method for the assay of beta activity by evaporating large water samples to dryness and counting the residue (Wheler, Kaufman, and Eliassen, 1952) has been developed which will detect and estimate I^{131}, P^{32}, and S^{35} in concentrations of the order of 10^{-6}uC (micro curies) per sample. This method suffers primarily from the standpoint of the length of time required to evaporate as much as a gallon of water without spattering or spillage.

Lacy (Water and Sewage Works, 1955) describes a method whereby fission products are separated from solution by coagulation of ferric chloride with limestone. It is reported that the removal of fission products containing barium, cerium, cesium, ruthenium, and strontium

was in the range of 51--59 per cent. This method is hardly feasible for quantitative determination. Lauderdale (Tech. Inform. Service, 1952) describes a method employing $CaCO_3$, $Ca(OH)_2$ and CO_2 which is reported to remove up to 99.9 per cent of strontium. No data for other ions are listed.

Applying specifically to the problem of analysis of water for fallout from nuclear explosions, Hahn, Straub, and Conrad (J. Am. Water Works Assn., 1955) have described in detail the results of a survey of several analytical methods. They also attempted evaporative procedures and abandoned the method for the same reasons already listed. A method using ion exchange resin is described in which ten liters of Oak Ridge tap water containing twenty milligrams of Sr^{++} carrier and $Sr^{90}_{0} - Y^{90}$ tracer were passed through a column at a rate of two milliliters per minute. The column was eluted with 300 milliliters of 2.5 M HCl at the same rate, and the eluate evaporated. The method was deemed impractical for the following reasons:

1. A period of several days is required for the absorption and elution of the activity.

2. The eluted solution must be concentrated by evaporation.

3. Interfering ions are concentrated as well as the desired radioactive materials.

The authors proceed to describe a method based on directly precipitating the fission products as the oxalate or carbonate.

A comprehensive list of apparatus including ion chambers, electroscopes, internal proportional counters and continuous monitoring equipment, all suitable for use in radiological monitoring, together with a good description of each, how each operates, the manufacturers and current prices, is given in a Task Group Report to the Journal of the American Water Works, 1956.

III. ION EXCHANGE RESINS, PROPERTIES, AND USES

Two excellent books covering ion exchange principles and uses in analysis have recently appeared (Kunin and Myers, 1950; Samuelson, 1953) in addition to several excellent reviews of the literature (Anal. Chem., 1950, 1951, 1952, 1954, 1956) including theory, practices, and history of ion exchange. Especially noteworthy is the paper of Bauman and Eichhorn (J. Am. Chem. Soc., 1947) covering the properties of Dowex-50, the resin used in all work contained in this thesis.

CHAPTER III

EXPERIMENTAL PROCEDURE

I. PREPARATION OF THE RESIN

Method I. One-half pound of Dowex-50 resin was placed in a column 1 m. long and 5 cm diameter and back-washed with tap water until approximately one-fourth of the resin (the finer particles) had been washed over the top of the column. This removal of the fine particles helps reduce close packing in the resin column and increases flow rate, in addition to eliminating the discharge of the fines during analysis which would result in reduced recovery. The resin was then allowed to settle and 2 l of 1 M Na_2SO_4 was passed through the column under a positive pressure of 5 psi to fully convert the resin to the sodium form. The Na_2SO_4 was followed by 2 l of 6 M HCl to convert the resin back to the hydrogen form, and the resin was then washed with distilled water until the wash water showed no chloride upon treatment with silver nitrate solution. Two hundred mls of acetone were passed through the column to remove the water, and followed by dry air for one minute. The resin was then removed from the column and stored in a stoppered glass bottle.

Method II. One-fourth pound of resin was converted to the sodium form by stirring with 1 M Na_2SO_4, converted back to the hydrogen form with 6 M HCl, and washed with distilled water until chloride free. The resin was then washed with 100 ml of acetone to remove water, filtered with suction, and stored in a glass bottle. During the process of cycling and washing the resin, only enough time was allowed for the heavier resin particles to settle before the supernatent solution and fine resin particles were removed. By the time the resin was washed with acetone, practically all the resin particles settled in ten seconds.

Both methods of resin treatment have been used by other workers in the past and seem to give the same satisfactory results. In the rest of this paper no distinction will be made between experiments on the basis of method of resin classification used.

II. DETERMINATION OF PER CENT ASH IN RESIN

Varying amounts of unclassified resin, i.e., as received from the manufacturer, were weighed into 25 ml porcelain crucibles and heated over a Meker burner for thirty minutes, cooled, weighed, and refired until successive heatings resulted in less than one mg loss in weight. It

was found that for a resin weight of ten grams approximately forty-five minutes heating period was sufficient. The resin weights and per cent ash are given in Table I.[1]

III. EXPERIMENTS WITH STRONTIUM NITRATE SOLUTION

A 0.05 M solution of $Sr(NO_3)_2$ was made up by dissolving 10.583 grams of $Sr(NO_3)_2$ (anhydrous) in a one liter volumetric flask. Thirty milliliters of this solution contained 0.33 g of strontium nitrate or 137 mg of Sr^{++}.

The apparatus for the preparation of the column is pictured in Figure 1.[2] To fill the column a plug of surgical cotton is placed in the column at A, and the column connected with rubber tubing to the overflow tube B. The column is filled with distilled water through the separatory funnel C until water flows from the overflow tube. The resin is then slurried with distilled water, poured into the funnel and allowed to settle under its own weight, displacing water through the overflow. Note that the bend of the overflow tube is high enough to keep a layer of water over the resin at all times. After all the

---

[1] Tables I through IX, see Appendix, pages 28-36.

[2] Figure 1 through 5, see Appendix, pages 37-41.

resin has settled into the column, the separatory funnel is removed and replaced by a 1 l filter flask (Figure 2). The strontium solution, containing 30 ml of the strontium nitrate stock solution in 500 ml of distilled water, is placed in the flask, the stopper wired down, and a positive five psi of air pressure applied through the flask side arm, D. Just as the first bubbles begin to appear in the delivery tube E, the pressure is relieved, and by means of a curved-stem funnel 200 ml of acetone are introduced into the flask and the procedure repeated, this time, however, allowing all the liquid to run through the column and following it with a stream of air for about a minute.

Since the concentration of the strontium solution was 137 mg Sr^{++} in 500 ml of water, or 2.7×10^{-4} grams per ml, and approximately 20 ml remained in the flask when diluted with 200 ml of water, the concentration after diluting is then 2.7×10^{-5} grams Sr^{++} per ml and similarly 100 ml of acetone dilutes this by a factor of 5:1. The final concentration of the 10 ml of solution remaining is then 0.5×10^{-5} grams Sr^{++} per ml. This is a total amount of Sr^{++} equal to 5×10^{-6} grams of Sr^{++} which constitutes a negligible loss.

After passing the acetone and air through the column, the resin and the cotton plug are extruded directly

into a tared crucible by pushing the plug from the bottom with a stirring rod. Some care is required in selecting the size for the plug. If the plug is too large it will retard the flow rate, if too small, much resin will be lost through adhering to the column walls during removal. It was found that a plug from 0.1-0.2 grams was suitable, the ash content of such a plug being negligible.

The ashing of the resin is accomplished by first igniting the acetone-wet resin, allowing it to burn quietly until it extinguishes itself. The crucible is then heated gently at first, finally being brought up to a red heat and kept there for at least twenty minutes. The crucible is cooled and weighed.

A control column is run along with the analysis in exactly the same manner but with no Sr^{++} solution. The weight of the control ash is subtracted from the weight of the analysis ash, and since the resin is a sulfonated polystyrene, ash is in the form of strontium sulfate, and the difference is expressed as grams of strontium. The results are listed in Table II.

IV. PROCEDURES USING $Th(No_3)_4$

Column Process

Since the reproductibility of analyses with strontium was good, but however the per cent recovery was low, it was felt that better techniques were required to improve the recovery. Since Th^{232}, the natural element, duplicates closely the chemistry of the fission products, and since it can be estimated quantitatively by its natural radioactivity, it was decided to make use of a $Th(NO_3)_4$ solution for further work in order to eliminate the tedious gravametric procedures.

A solution containing 10.3 g of anhydrous $Th(NO_3)_4$ was made up in a 500 ml volumetric flash. Since the resin when wetted with acetone might present a difficulty in weighing accurately, a portion was allowed to air dry over night and one column was made with this dried resin and another with the acetone-wet resin. Using 15 ml of the $Th(NO_3)_4$ solution the procedure was exactly that described for strontium. In addition, a sample containing 15 ml (153 mg Th^{++++}) of the Th^{++++} solution was carefully evaporated in a crucible at never more than 80°C and the radioactivities of all three were measured using an end window Geiger-Muller tube and a Berkeley Decimal Scaler. The results are given in Table III.

Batch Process

Several determinations were made in which a batch process instead of a column method was employed. The procedure was to pipette 15 ml (153 mg Th^{++++}) of the $Th(NO_3)_4$ solution into a 125 ml Erlenmeyer, dilute with 100 ml distilled water and add 5 g of unclassified resin weighed on a platform balance. The mixture was stirred by a magnetic stirrer for at least fifteen minutes and the resin filtered out, washed with 25 ml of acetone, and allowed to dry for about one minute. The filter paper was carefully wrapped around the resin and the whole ashed as before. In addition, the filtrates were carefully evaporated in an oven at 80°C, washed into a crucible identical with those used for ashing, evaporated to dryness and the radiation counted. A blank was run with no thor (filtrate nor evaporated); 15 ml of the thorium nitrate solution was evaporated and counted as before. The results are given in Table IV.

To determine the effect of high dilution on recovery, a 15 ml Th^{++++} sample was diluted to 4 l and run exactly as the other batch processes. A similar amount of $Th(NO_3)_4$ was run in 4 l of distilled water containing 500 ppm (2 g) of NaCl. A portion of the filtrate from this run was evaporated to dryness and counted. (Table IV)

Estimation of the amount of self absorption in $Th(NO_3)_4 \cdot 12\ H_2O$. Since evaporated samples of $Th(NO_3)_4$ solutions consistently count appreciably lower than similar samples recovered by ion exchange, and further since the resin shows no activity by itself (Tables III and IV) the low counts on evaporated samples can be ascribed only to self absorption of the alpha rays by water of crystallization in the thorium nitrate. From aqueous solution thorium nitrate crystallizes as $Th(NO_3)_4 \cdot 12\ H_2O$, this was not realized when Tables III and IV were prepared. In order to determine a factor for correcting the activity of a $Th(NO_3)_4 \cdot 12\ H_2O$ sample to the activity of an anhydrous $Th(SO_4)_2$ residue such as derived from column or batch operations, three samples of 15 ml of $Th(NO_3)_4$ solution were evaporated in the oven at 80°C and counted. These were then heated over a Meker burner for ten minutes (with evolution of H_2O and NO_2) and allowed to cool. Five drops of concentrated H_2SO_4 were then added, the samples carefully evaporated to dryness and counted again. The results appear in Table V.

Determination of effect of inactive salts in water samples. Since the previous experiments led to encouraging results, it was decided to attempt to apply the procedure to solutions containing a high percentage of dissolved

solids (such as NaCl). This condition will occur in most domestic water supplies and will be expected to necessitate both the use of larger amounts of resin and result in a reduction of apparent activity due to self absorption losses.

To determine the amount of resin necessary for absorption of all the thorium from a sample containing other dissolved solids, solutions containing 153 mg of Th^{++++} and 2 g (500 ppm) of NaCl were made up in 4 l of distilled water. These solutions were passed through columns containing 1, 2, 4, 8, and 16 grams of resin, respectively, a fifth solution was directly evaporated to dryness and all residues were counted. It was found that the deployment of the residue in the crucible had no effect on the counting rate. The results are given in Table VI and plotted in Figure 3.

The extent of self absorption of radiation by foreign salts in a sample was determined by employing solutions containing 153 mg of Th^{++++} and various concentrations of NaCl, representing a range of water hardness such as may be encountered in domestic water supplies. A 30 per cent solution of NaCl was prepared and quantities from one to ten mls were pipetted into crucibles idential to those used for ashing. Next, 153 mg of Th^{++++} was added

(as 15 ml of $Th(NO_3)_4$ solution) and the volume in all crucibles made up to 25 ml with distilled water. Each ml of the 30 per cent NaCl solution contained a weight of salt equal to the weight of sodium chloride contained in a 4 l sample of 75 ppm dissolved solids expressed as NaCl. The contents of each crucible were evaporated to dryness, ignited over a Meker burner until all obvious reaction ceased, cooled and counted. Results are given in Table VII and in Figure 4.

V. EXPERIMENTS WITH FISSION PRODUCTS

The fission products were obtained from the chain reacting pile at Oak Ridge, Tennessee, and contained 0.0109±5 per cent mc/ml of mixed fission products as nitrates in a nitric acid solution of volume 0.1 ml. Total solids were quoted by the A. E. C. as 21 mg/ml and non volatile materials as 21 mg/ml. They were further described as "mixed fission products from heavy metal irradiations less than forty-five days old." Table VIII gives the expected fission products, their half lives and types of radiation.

(References on Table VIII). The 0.1 ml of fission products was made up to 100 ml in a volumetric flask, and from this flask 10 ml aliquots were taken and further diluted to 100 ml to be used in the experiments.

10 ml of the fission product solution (containing approximately 200 counts/ml) were added to four liters of distilled water, the solution run through a column containing 5 g of Dowex-50 as before, and the resin ash counted. One-half of the resulting effluent was evaporated to dryness and counted, while the other 2 l portion was run through a second column containing 5 g of Dowex-50, and the ash from this second resin column was counted. Finally, a batch process was run using 10 ml of fission products in 4 l of distilled water and stirred for at least fifteen minutes before filtering and ashing. The results are given in Table IX.

CHAPTER IV

METHODS OF CALCULATIONS AND DISCUSSION OF RESULTS

I. METHODS OF CALCULATIONS

Method of calculating the standard deviation.

Standard deviations on counting rates are calculated by the equation $\sigma = \sqrt{N}$, where N is the total number of counts. The following formulae have also been used for calculating the standard deviations on the sums, differences, and quotients of counting rates:

Sums: $$\sigma_s = \sqrt{\sigma_1^2 + \sigma_2^2 + \dots}$$

where is the standard deviation of the sum of the counting rates whose deviations are σ_1, σ_2 etc.

Differences: $$\sigma_d = \sqrt{\sigma_1^2 + \sigma_2^2}$$

where is the standard deviation on the difference of the counting rates whose deviations are σ_1 and σ_2.

Quotients:

If the divisor does not have a standard deviation on it, the dividend and its standard deviation are both divided separately. For example:

$$\frac{N \pm \sigma}{t} = \frac{N}{\tau} \pm \frac{\sigma}{t}$$

If the divisor also has a standard deviation, the expression for the standard deviation on the quotient becomes:

$$\frac{\sigma_Q}{Q} = \sqrt{\frac{\sigma_1^2}{R_1^2} + \frac{\sigma_2^2}{R_2^2}}$$

where σ_Q is the standard deviation on the quotient, Q, and σ_1 and σ_2 are the standard deviations on counting rates R_1 and R_2 respectively.

It should be pointed out that in all tables the standard deviation has been calculated by the above formulae and are therefore standard deviations due only to the randomness of radioactive disintegration. In no way do they represent standard deviations on analytical techniques, variations in counter geometry or voltages, or any other factors within the control of the experimenter. Since these sources of error cannot be shown to be negligible, it must be clearly understood that all standard deviations represent a minimum rather than a true measure of experimental error.

Calculations of the absorption factor for $Th(NO_3)_4 \cdot 12H_2O$ from the data in Table V are given at the bottom of the table.

Per cent self absorption given in Table VII was calculated by dividing the activities of the various residues containing sodium chloride by the activity of the

evaporated solution containing no sodium chloride and multiplying by 100.

II. DISCUSSION OF RESULTS

It is interesting to note that more or less complete conversion of strontium to the sulfate form by the resin was accomplished. If this method were to be applied to other ions probably a few drops of sulfuric acid should also be added when ignition is almost complete to ensure complete conversion of the ion to the sulfate form. The 81 per cent recovery was somewhat low as ion exchange processes go, but in the series for absorption of various ions in resins given earlier in this paper, strontium is near the weaker end of the series. Aslo, Hahn, Straub, and Conrad (J. Am. Water Works Assn., 1955) found only an 80 per cent recovery of strontium by an ion exchange process from an Oak Ridge tap water solution they had prepared containing 30 mg of Sr^{++} carrier and $Sr^{90} - Y^{90}$ tracer. They eluted the column with HCl and precipitated strontium as the oxalate. They attributed the loss to calcium contamination of the final strontium oxalate precipitate, rather than actual loss due to incomplete removal by the resin. Since the sample was run in distilled water, the experiments described in this paper seem rather to indicate

that strontium does not absorb completely on an ion exchange resin.

Since thorium is a weak alpha emitter (α - 4.30 mev), serious self absorption effects were expected. That these effects were found can be seen by the absorption of 30 per cent of the activity by water of crystallization in the thorium nitrate as compared to the anhydrous thorium sulfate. As might be expected, foreign salts in the sample were found to increase self absorption to a high degree, and the plot of Figure 4 shows that absorption apparently follows exponential curves, with residual activity being due to beta and gamma radiation from thorium daughter products such as Ra^{228}, Ac^{228}, and Th^{228}.

A combination of Figures 3 and 4 yields a three dimensional plot, Figure 5, with coordinates of activity, weight of resin and parts per million dissolved solids. Data are not sufficient to draw the complete graph, sufficing however for one complete curve and one point on each of the other five curves. The dotted lines indicate the probable configurations of these curves.

Table IX reveals the presence of a problem with the actual fission products that was not encountered in the preliminary experiments with thorium. The total activity present in the 4 l sample treated by the column method

was 1913±14 counts per minute. Of this amount, 1020±10 counts per minute were removed by the resin column the first time. Multiplying the activity of one-half the effluent by two gives a total activity remaining in the effluent of 728±12 counts per minute. The sum of these two gives 1748±16 counts per minute, which subtracted from 1913±14 leaves 165±21 counts per minute still to be accounted for. It is thought that this activity is due to self absorption in either the effluent residue, or more probably, by the resin ash from the column operation. Even so the fact remains that the resin column only removes about one-half the total activity even when run with distilled water. Of the 728±12 counts remaining in the effluent, only 128±8 can be removed by treating with resin a second time, indicating that the material must be in some form other than cationic. This form may well be colloidal and perhaps could be removed by passing the sample down a column containing both resin and activated charcoal. An alternative explanation is that the material is anionic and could be efficiently picked up by using a mixed bed column (i.e., one composed of both anion and cation exchange resins).

Absorption of the radiation by the presence of foreign ions as would be encountered in tap water might well be reduced by precipitating these ions. Calcium and

magnesium would be expected to be the most troublesome ions, but experience has shown that treating a tap water sample with a little sodium carbonate solution will, on standing, produce a precipitate of magnesium carbonate and calcium carbonate. The precipitate is colloidal in nature and will pass through ordinary coarse filter paper and at least partially through a cation resin column as well, thus eliminating a large quantity of interfering ions. Of course, sodium carbonate could not be used as the sodium would contaminate the resin and nothing would be gained. However, if ammonium carbonate were employed, sufficient resin could be used to absorb all ions, and during ashing the ammonium salts would be volatilized.

Time limitations required that the research be terminated at this point, however the author believes that the procedure is an improvement over the evaporative method, and perhaps the suggestions made above will in time lead to refinements of this procedure by reducing self absorption of the activity and improving recovery by the resin, resulting in a still more rapid and accurate method for estimation of radioactivity in public water supplies.

BIBLIOGRAPHY

BIBLIOGRAPHY

A. BOOKS

Friedlander, G. and J. Kennedy. Introduction to Radiochemistry. New York: John Wiley & Sons, 1949.

Kunin, Robert, and Robert J. Myers. Ion Exchange Resins. New York: John Wiley & Sons, 1950.

Samuelson, Olof. Ion Exchangers in Analytical Chemistry. New York: John Wiley & Sons, 1953.

B. PERIODICALS

Ayres, John A. "Treatment of Radioactive Waste by Ion Exchange," Ind. Eng. Chem., 43:1526-31, 1951.

Bauman, W. C., and J. Eichhorn. "Fundamental Properties of a Synthetic Cation Exchange Resin," J. Am. Chem. Soc., 69:2830-36, 1947.

Benedict, J. T., Walter C. Schumb, and Charles D. Corvell. "Distribution of Zirconium and Hafnium Between Cation Exchange Resin and Acid Solutions, The Column Separation with Nitric Acid--Citric Acid Mixture," J. Am. Chem. Soc., 76:2036-40, 1954.

Beukenkamp, John, and William Reiman III. "Determination of Sodium and Potassium Employing Ion Exchange Separation," Anal. Chem., 22:582-85, 1950.

Brown, William E., and William Rieman III. "The Separation of Titanium, Zirconium, and Thorium by Ion Exchange," J. Am. Chem. Soc., 74:1278-82, 1952.

Downing, A. L., A. B. Wheatland and G. E. Eden. "The Removal of Radioisotopes During the Treatment of Domestic Water Supplies II Radiostrontium," J. Inst. Water Engrs., 7:555-72, 1953.

Freiling, Edward C., and Leland R. Bunney. "Ion Exchange as a Separations Method. VII. Near Optimum Conditions for the Separation of Fission Products Rare Earths with Lactic Acid Elution at 87°," J. Am. Chem. Soc., 76:1021-22, 1954.

Hahn, Richard B., and Conrad P. Straus. "Determination of Radioactive Strontium and Barium in Water," J. Am. Water Works Assn., 47:335-49, 1955.

Hudgens, Jame E., Jr. "Analytical Applications of Radiochemical Techniques," (Review), Anal. Chem., 24:1704-8, 1952.

Ketelle, B. H., and G. E. Boyd. "Further Studies of the Ion Exchange Separation of the Rare Earths," J. Am. Chem. Soc., 73:1862-63, 1951.

Kilcawleu, E. J., and others. "Measurement of Radioactive Fallout in Reservoirs," J. Am. Water Works Assn., 46: 1901-2111, 1954.

Kunin, Robert. "Ion Exchange," (Review), Anal. Chem., 23: 45-6, 1951.

_____. "Ion Exchange," (Review), Anal. Chem., 24:64-5, 1952.

_____, and Francis X. McGarvey. "Ion Exchange," (Review), Anal. Chem., 26:104-9, 1954.

Lacy, William J. "Removing Radioactive Materials from Water by Coagulation," Water and Sewage Works, 100:410-11, 1953.

Lauderdale, Robert A. "Decontamination of Radioactive Water," U. S. Atomic Energy Comm., Tech. Inform. Service, Oak Ridge, Tenn., Wash - 129, (1-4), 1952.

Marinsky, Jacob A. "A Study of the Ion Exchange Equilibria Between Hydrogen Ions and Sodium, Calcium, Barium, and Cerium Ions," Tech. Report No. 34. Laboratory for Nuclear Science and Engineering, M. I. T., Cambridge, Mass., 1949.

Nader, J. S., A. S. Goldin, and L. R. Setter. "Radioactive Fallout in Cincinnati Area," J. Am. Water Works Assn., 46:1096-1100, 1954.

Schubert, Jack. "Analytical Applications of Ion Exchange Separations," (Review), Anal. Chem., 22:1359-68, 1950.

Straus, Conrad P. "Water Decontamination Studies--Oak Ridge National Laboratory," U. S. Atomic Energy Comm., Tech. Inform. Service, Oak Ridge, Tenn., Wash. - 139, 84-89, 1952.

Task Group Report, "Instrumentation for Radioactive Pollution Studies," J. Am. Water Works Assn., 48:397-416, 1956.

Tompkins, Edward R. "Ion Exchange Separations," (Review), Anal. Chem., 22:1352-59, 1950.

_____, J. X. Khym, and W. E. Cohn. "Ion Exchange as a Separations Method. I The Separation of Fission-Produced Radioisotopes, Including the Individual Rare Earths, by Complexing Elution from Amberlite Resin," J. Am. Chem. Soc., 69:2769-77, 1947.

Wheelwright, E. J. and F. H. Spedding. "A Rapid Method for Fractionating Crude Rare Earth Ores into Mixtures Greatly Enriched with Regard to Particular Rare Earths," J. Am. Chem. Soc., 75:2529-80, 1953.

Wheler, Gordon A., Jr., Warren J. Kaufman and Rolf Eliassen. "Assaying Techniques for Radioisotopic Contaminations in Water Supplies," U. S. Atomic Energy Comm. NYO-4437, 3-79, 1952.

APPENDIX

TABLE I

ASH CONTENT OF UNCLASSIFIED DOWEX-50 ION EXCHANGE RESIN

Weight of Resin in Grams	Weight of Ash in Grams	Per cent Ash
1.08	0.001	0.09
1.81	0.003	0.17
7.99	0.009	0.11
9.37	0.010	0.11
14.11	0.017	0.12
	Average	0.12

TABLE II

RECOVERY OF STRONTIUM BY DOWEX-50 COLUMN PROCEDURE

Sample	Resin Wt.	Wt. Ash	Wt. $SrSO_4$	Wt. of Sr Recovered	Wt. of Sr Introduced	Per cent Recovery
Control I	7.994	0.016	____	_____	_____	_____
Anal. I	7.998	0.243	0.227	0.109	0.137	79.5
Control II	6.000	0.010	____	_____	_____	_____
Anal. II	6.000	0.246	0.236	0.113	0.137	81.0

TABLE III

RECOVERY OF THORIUM BY DOWEX-50; COLUMN PROCEDURE

Sample	cpm ± σ		
	Sample	Background	Net cpm
Dry Resin column (3.05 g)	1217 ± 11	30.3 ± 0.58	1187 ± 11
Acetone-wet Resin column (6.00 g)	1187 ± 11	30.3 ± 0.58	1157 ± 11
Evaporated	950 ± 10	30.3 ± 0.58	920 ± 10 (corrected to 1200 ± 13)*

*Factor from Table V.

TABLE IV

RECOVERY OF THORIUM BY DOWEX-50; BATCH PROCEDURE, 5g UNCLASSIFIED RESIN

Sample	cpm±σ		
	Sample	Background	Net
I - 153 mg Th stirred	1470±12	49.9±2	1420±12
II - 153 mg Th stirred	1504±12	81.4±3	1423±12
5 g resin no Th	46.5±2	48.5±1.7	———
Filtrate from I	86.5±3	46.3±3	40.2±4.2
Filtrate from II	96.5±3	46.3±3	50.2±4.3
153 mg Th stirred 4 L	1063±10	38.1±2	1025±10
153 mg Th stirred in 4 L 500 ppm NaCl	610±8	52.1±2	558±8
sol'n containing 153 mg Th-evap.	1110±10	46±3	1064±11 (corrected for self absorb. 1382±14)*

*Factor from Table V.

TABLE V

COMPARISON OF $Th(NO_3)_4 \cdot 12\,H_2O$ AND $Th(SO_4)_2$ ACTIVITIES

Sample	cpm ± σ			
	Sample	Background	Net	#
I - 15 ml Th sol'n. containing 153 mg Th evap.	1086±10	33.1±1.8	1053±11	1
II - 15 ml Th sol'n. containing 153 mg Th evap.	1072±10	43.7±2.1	1028±11	2
III - 15 ml Th sol'n. containing 153 mg Th evap.	1093±10	49.8±2.2	1043±11	3
I converted to sulfate	1417±12	33.1±1.8	1384±12	1a
II converted to sulfate	1319±12	43.7±2.1	1347±12	2a
III converted to sulfate	1426±12	49.8±2.2	1376±12	3a

Calculation of conversion factor $\frac{Th(SO_4)_2}{Th(NO_3)_4 \cdot 12H_2O}$

1a/1 -- 1.3±0.02
2a/2 -- 1.3±0.02
3a/3 -- 1.3±0.02

TABLE VI

RECOVERY OF THORIUM FROM 4 l OF WATER CONTAINING 500 ppm SODIUM CHLORIDE

Sample	cpm ± σ		
	Sample	Background	Net
Ash from 1 gram resin	145±3.6	62±1	81±4.4
Ash from 2 grams resin	302±3.1	62±1	237±3.8
Ash from 4 grams resin	421±6	62±1	341±7
Ash from 8 grams resin	587±8	62±1	520±8
Ash from 16 grams resin	645±8	62±1	582±8
Evaporated sample	450±6	64±2	386±6

TABLE VII

EFFECT OF WATER HARDNESS ON RECOVERY OF THORIUM ACTIVITY

ppm sodium chloride	cpm±σ			% Self-absorp.
	Sample	Background	Net	
0	1210±10	53±4	1157±11	0
75	871±9	64±5	808±10	30.0±1
150	736±9	48±4	699±9	39.6±1
225	652±8	43±4	609±9	47.3±1
300	560±8	48±4	512±8	55.6±1
375	553±8	47±3	506±8	56.1±1
450	536±7	41±4	495±8	57.2±1
525	507±5	48±3	459±6	60.3±1
600	502±6	44±4	458±8	60.4±1
675	484±7	45±3	439±8	62.0±1
750	469±7	42±3	427±8	63.0±1

TABLE VIII***

HAZARDOUS URANIUM FISSION PRODUCTS

Fission Product	Half Life	Type of Radiation and Energy*
Sr^{89}	54 d	B^- 1.50 no γ
Y^{91}	57d	B^- 1.53 no γ
Zr^{95}	65d	B^- 0.394, 1.0; γ 0.73, 0.92?
Nb^{95} (Cb^{95})	90h	IT 0.24; e^- 0.22, 0.24
Ru^{106}	1.0 y	B^- 0.03; no γ
Rh^{106}	30 s	B^- 3.55, 2.30; γ 0.51, 0.73, 1.25
I^{131}	8 d	B^- 0.595, 0.315; e^- 0.080; γ 0.363, 0.638, 0.283
Cs^{137}	33 y	B^- 0.550
Ba^{137}	2.6m	IT 0.663; γ 0.663; e^- 0.626, 0.657
Ba^{140}	12.8d	B^- 1.05, 0.34?; γ 0.54; e^- 0.50
La^{140}	40h	B^- 1.4, 0.9, 2.12; γ 1.64, 0.85, 0.49, 2.4, 0.335
Ce^{144}	275d	B^- 0.348; e^- 0.075, 0.12; no γ
Pr** ^{144}	17.5m	B^- 3.07; e^- 0.091, 0.128, 0.103?; γ 0.135, 0.145?, 1.25

*Energy in million electron volts.

**All products except Pr^{144} and possibly I^{131} are undoubtedly present in the mixed fission products as received from the A. E. C.

***From Hahn, Straub, and Conrad (1955), and Friedlander and Kennedy (1949).

NOTE: IT--Isomeric transition
e^---Internal conversion electrons

TABLE IX

RECOVERY OF URANIUM FISSION PRODUCTS WITH DOWEX-50

Sample	cpm±σ		
	Sample	Background	Net
A - 10 ml mixed fission products solution directly evaporated	1963±14	50±2	1913±14
B - 10 ml mixed fission products solution in 4 L distilled water Column procedure	1074±10	54±2	1020±10
C - ½ effluent from "B" evap.	411±6	47±2	364±6
D - ½ effluent from B treated with 5 g resin in column	111±3	48±2	63±4
E - 10 ml mixed fission products solution in 4 L distilled water batch procedure	738±9	53±2	685±9

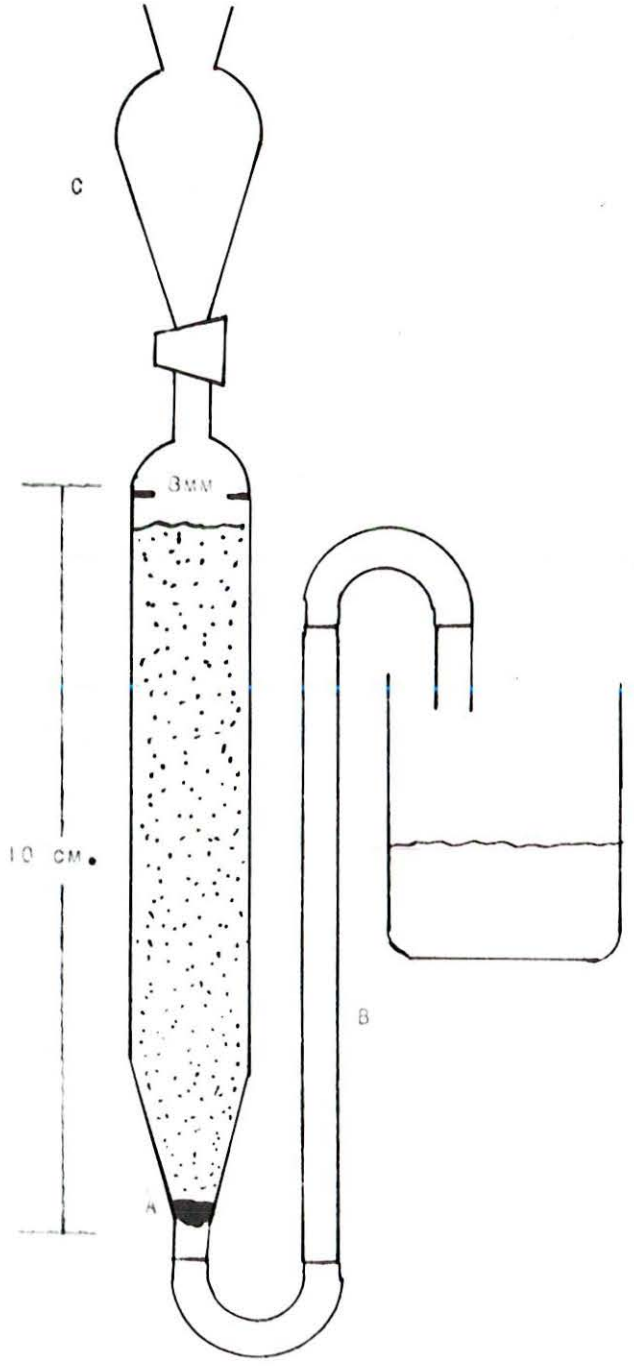


FIGURE 1

APPARATUS FOR PREPARATION OF THE COLUMN

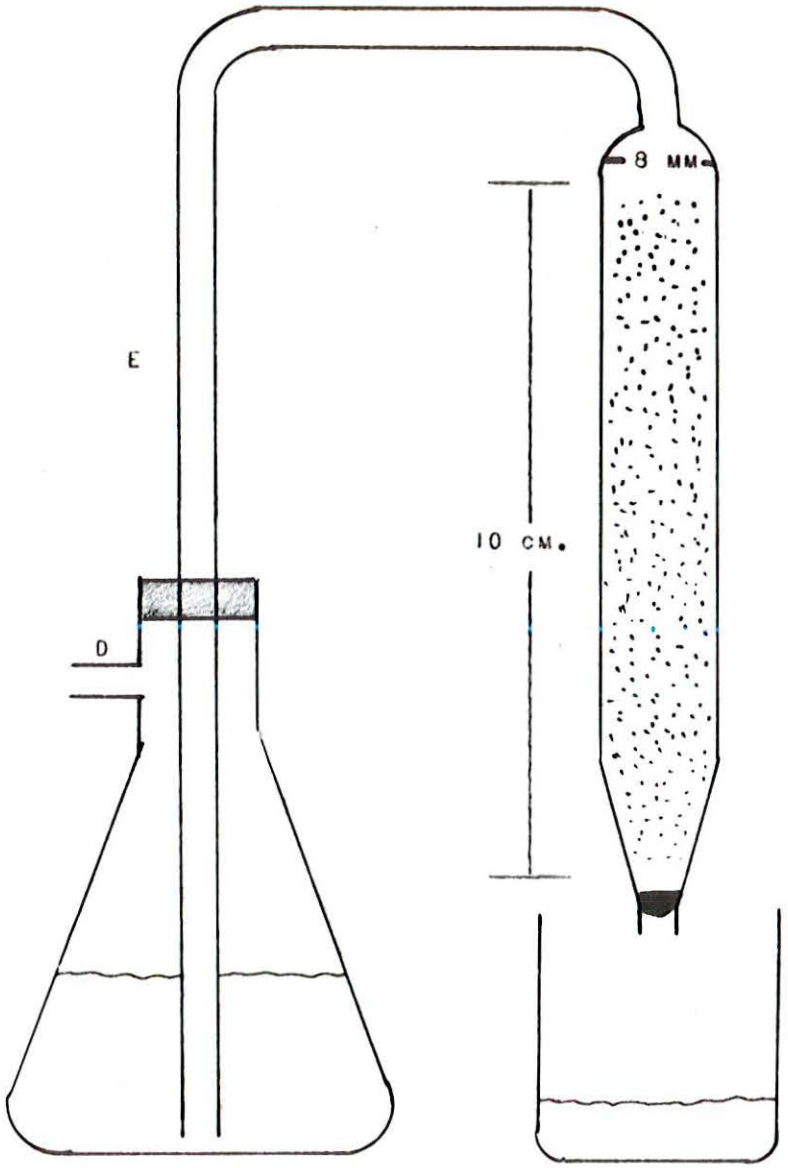
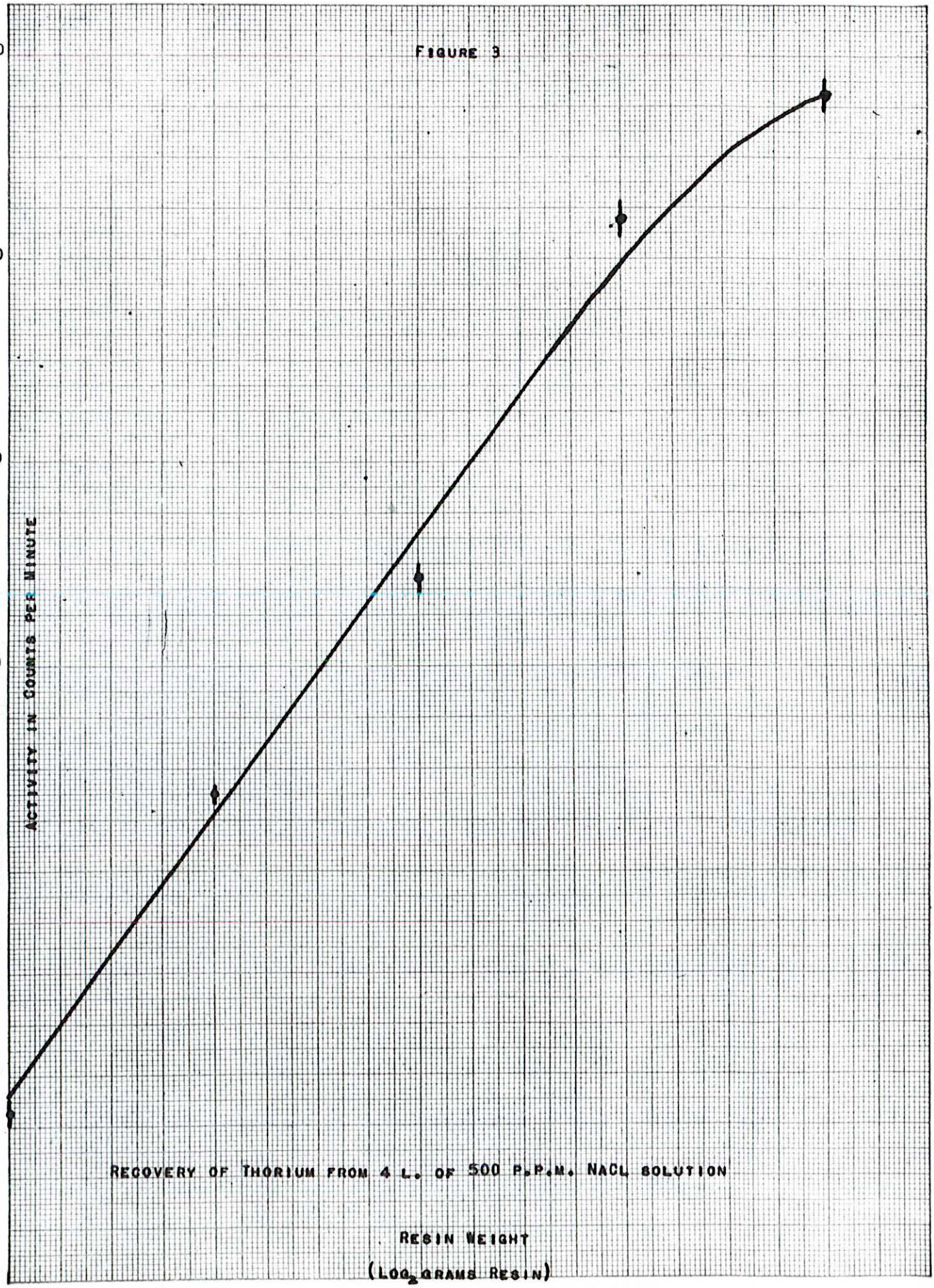


FIGURE 2

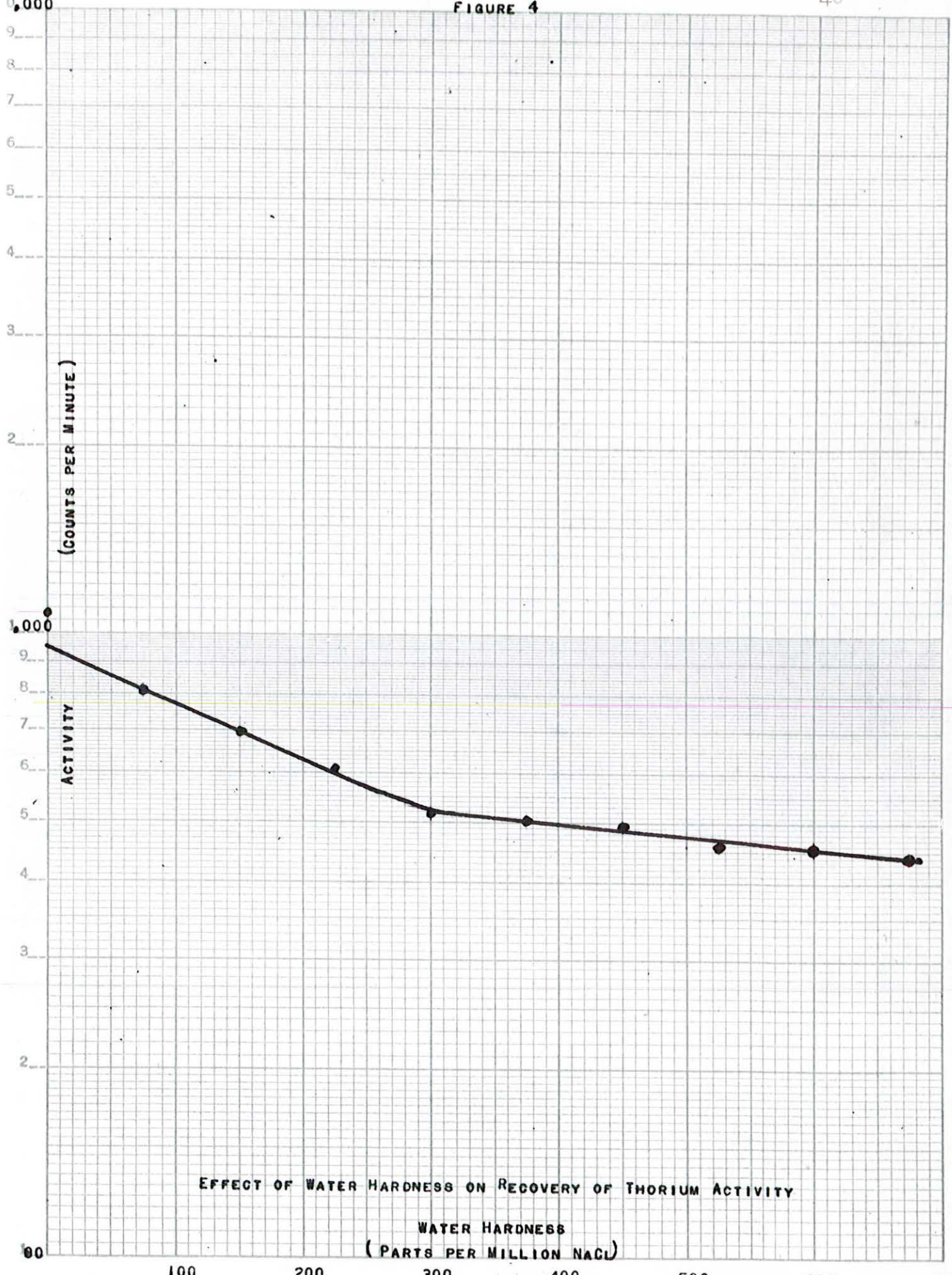
APPARATUS FOR COLUMN PROCEDURE

FIGURE 3



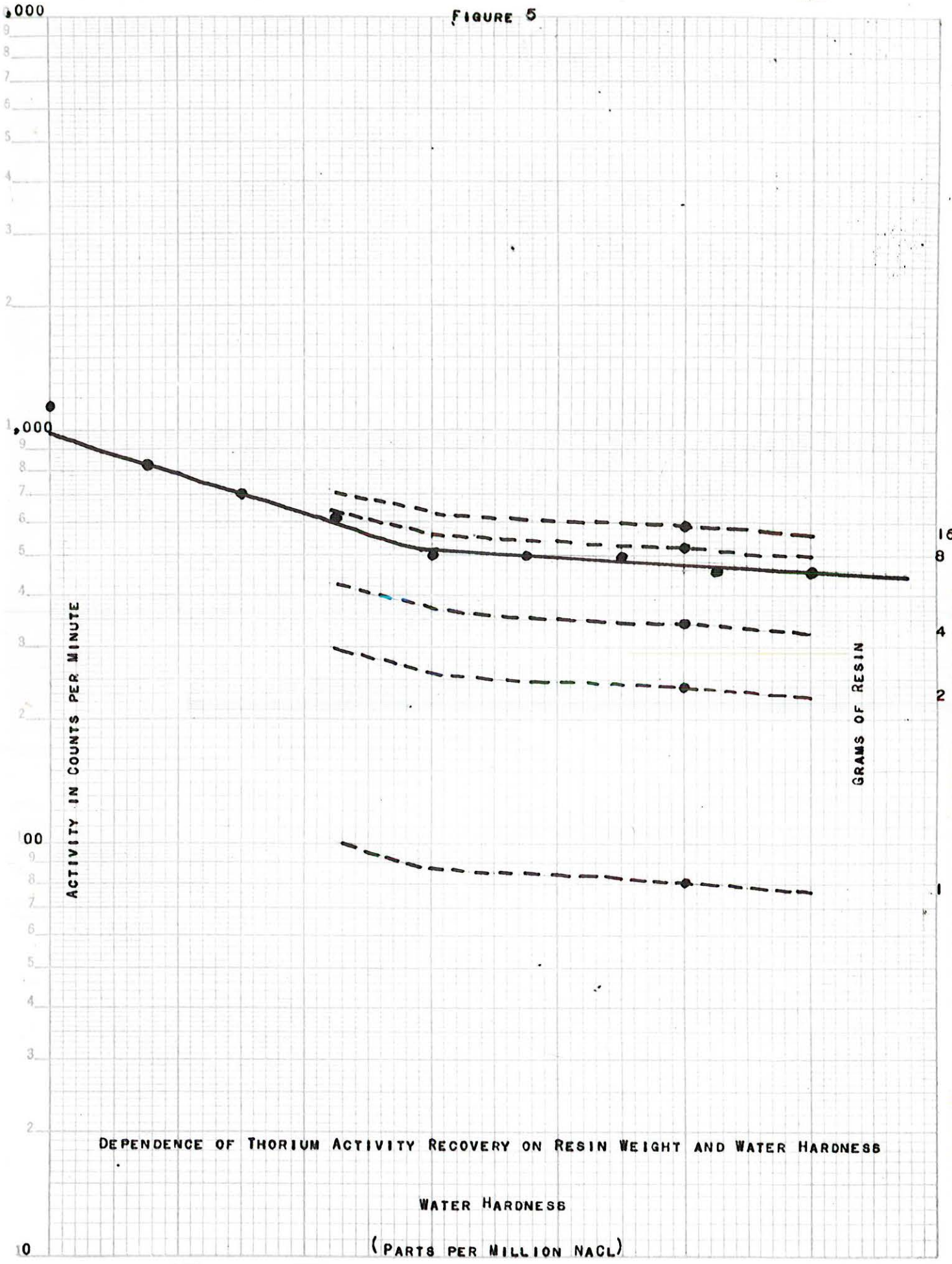
RECOVERY OF THORIUM FROM 4 L. OF 500 P.P.M. NACL SOLUTION

FIGURE 4



EFFECT OF WATER HARDNESS ON RECOVERY OF THORIUM ACTIVITY

FIGURE 5



DEPENDENCE OF THORIUM ACTIVITY RECOVERY ON RESIN WEIGHT AND WATER HARDNESS