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DEVELOPMENT OF A CONTINUOUS ION EXCHANGE PROCESS FOR THE REMOVAL AND RECOVERY OF HIGH-PURITY CESIUM FROM ALKALINE WASTE

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Chemical Development Section B

DEVELOPMENT OF A CONTINUOUS ION EXCHANGE PROCESS FOR THE REMOVAL AND RECOVERY OF HIGH-PURITY CESIUM FROM ALKALINE WASTE

I. R. Higgins, A. F. Messing

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ABSTRACT

A process was developed for the removal and recovery of high-purity cesium from alkaline waste solutions using the Higgins continuous countercurrent ion exchange contactor. Cesium is selectively sorbed on the phenolic methylene sulfonic resin Duolite C-3, along with strontium, the rare earths, and other cationic fission products. At feed rates up to 2000 gal/ft^2.hr, tracer activities in a synthetic alkaline waste solution were reduced to background levels of 0-50 c/ml.min, corresponding to decontamination factors of $>10^4$ for cesium and $>10^3$ for strontium and the rare earths. Cesium was removed from the contactor by selective elution with hydrochloric acid to produce a product containing about 1% sodium along with tracer quantities of strontium and the rare earths.

A cost estimate was prepared which showed that Cs-137 would cost 6 cents per curie from a plant designed to produce 22,000 curies/day.

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1.0 INTRODUCTION

This investigation was undertaken in order to develop a method for the efficient removal of cesium and other long-lived radionuclides from alkaline wastes of high salt content, this being, in effect, a step in a general waste-treatment process. A second objective was to recover the cesium in a highly pure form suitable for use as a gamma source.

With the increasing use of nuclear fuels for power production, the problem of waste disposal becomes one of increasing significance. At present, process wastes are generally neutralized with sodium hydroxide and stored in tanks or pits designed to contain the solution for long periods. After neutralization of the acid waste there remains in solution 0.1-1.0% of the original strontium, rare earths, americium, and plutonium along with most of the cesium and about 10% of the ruthenium. Because of the biological hazards[1] and the long half-lives of the species present, direct disposal of the waste solutions to the environment is limited. On the other hand, if these nuclides were removed from solution, the prospective storage problem would be minimized in that a shorter hold-up time would be required prior to disposal.

Cesium-137 is of interest in that it is considered to be potentially one of the best gamma radiation sources.[2] As a radiographic source Cs-137 is comparable to Co-60 and radiographically similar to devices producing x rays under a peak potential of 1,000 kv. It has a high-energy monochromatic gamma ray of 0.66 Mev, a long half-life, 33 years, and a theoretical specific activity[3] of 23 curies/g. Though Cs-137 has a high fission yield, its general application as a radiation source is dependent on its processing cost.

Continuous experiments were made in a 1.5-in.-dia Higgins continuous countercurrent ion exchange contactor charged with the phenolic methylene sulfonic ion exchange resin Duolite C-3. Preliminary experiments were conducted on fixed beds of the resin in order to obtain distribution coefficients necessary for the establishment of sorption and eluting conditions in the continuous equipment. Synthetic alkaline wastes containing tracer amounts of Cs-137, Sr-90, or Ce-144, this last representing the rare earths and americium, was used as feed while 3.0 M HCl was used as the eluting agent.

Ion exchange techniques have recently received increased attention as a means of removing cesium from alkaline waste solutions of high salt content. Miller and Kline[4] found that cesium is selectively sorbed by the phenolic groups of phenolic methylene sulfonic ion exchange resins. Barney[5] found a high degree of cesium removal from alkaline waste solutions by a cation exchange resin of this type, Duolite C-3. At Hanford,[6] synthetic wastes were decontaminated from cesium on a fixed ion exchange bed of Duolite C-3 by factors of 10^4-10^5. Other cesium concentration methods include co-crystallization with ammonium alum[7,8] and scavenging with metal ferrocyanides.[9]

High-level decontamination has yet to be demonstrated in a shielded facility. Though americium is considered to behave like the rare earths, the action of plutonium should be investigated separately.

The authors acknowledge the assistance of E. R. Eastwood in the operation of the continuous equipment, H. H. Carmichael in fixed-bed studies, and P. L. Robertson in preparing the cost estimate. The authors are also indebted to G. R. Wilson and W. R. Laing and staffs of the ORNL Analytical Chemistry Division for analyses performed.

2.0 PROCESS DESCRIPTION

A flowsheet for a proposed plant capable of producing 22,000 curies of Cs-137 per day has been prepared (Fig. 2.1). The process also acts as a step in a general waste treatment method. Feed is assumed to be the supernatant from sodium hydroxide--neutralized nitric acid wastes from the first-cycle solvent extraction of dissolved irradiated fuel elements. Such waste contains practically 100% of the cesium and aluminum of the original waste together with 0.1-1.0% of the rare earths, strontium, americium, and plutonium and about 10% of the ruthenium.

In the continuous equipment[10] (see appendix) solutions and resin are contacted countercurrently with the following operations taking place simultaneously: Cesium and other cationic materials are sorbed on the sodium form of a phenolic methylene sulfonic resin, Duolite C-3, and the feed solution is washed from the void volume of the resin with water. Hydrochloric acid, 3.0 M, is passed through the column to selectively elute first the sodium and then the cesium. The eluant is washed from the void volumes with sodium chloride solution, which also regenerates the sodium form of the resin necessary to prevent precipitation of aluminum hydroxide in the column. In a conventional fixed-bed operation the five steps would be carried out consecutively, or batchwise.

The feed waste effluent activity is reduced to background levels in cesium, strontium, and the rare earths. However, because a major portion of the ruthenium together with 100% of the nitrate will still be present, further processing will be required prior to environmental release. The fate of plutonium is yet to be investigated.

Hydrochloric acid was chosen as the eluant because cesium gamma sources are prepared as cesium chloride and the extra conversion step is avoided. The equipment is therefore constructed of Hastelloy C. With nitric acid as eluant, stainless steel could be used, but nitric acid attacks the phenolic ion exchange resin. Sulfuric acid could be used if the cesium is to be isolated by the alum process.[7,8]

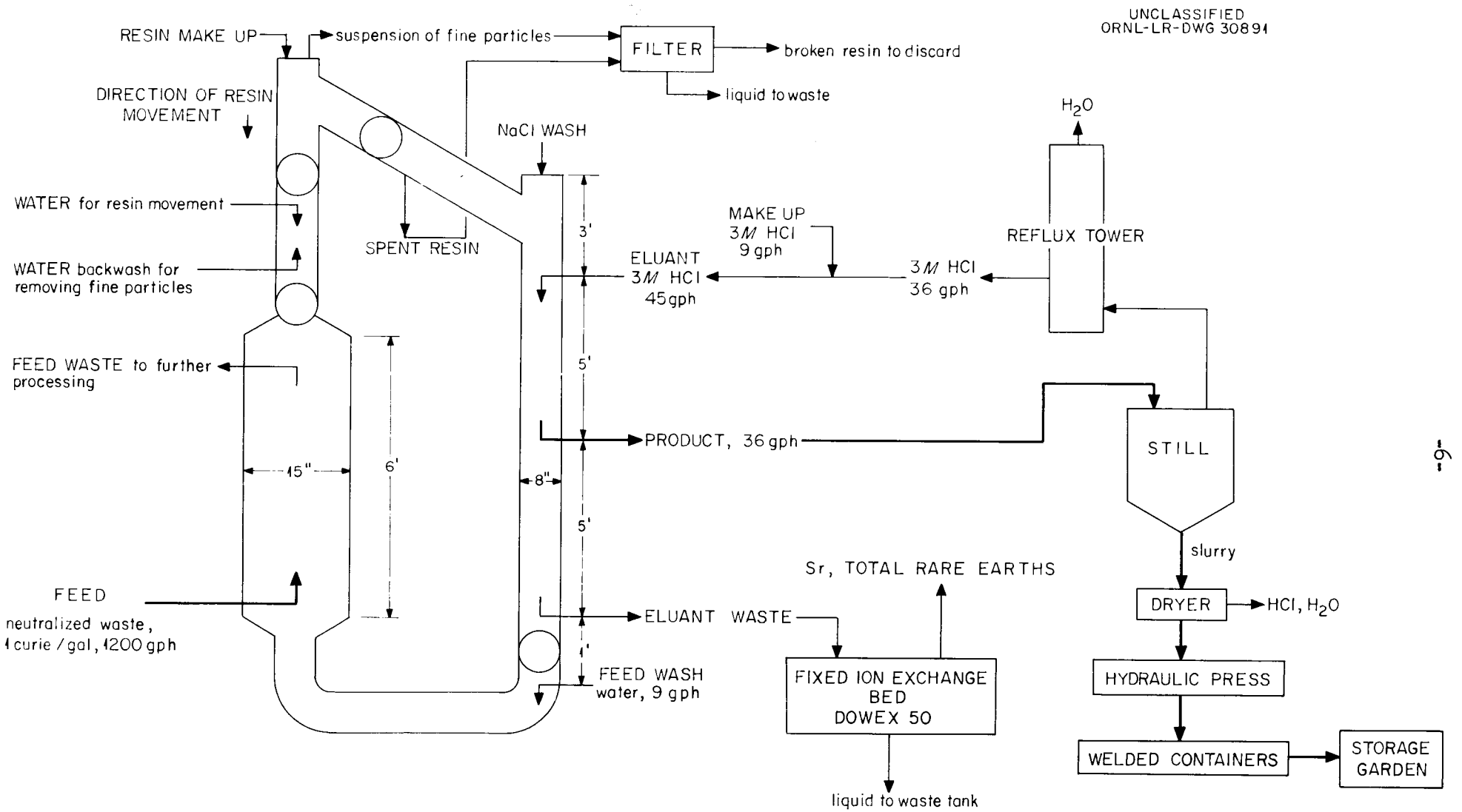

Fig. 2.1. Schematic Flowsheet for Waste Decontamination and for the Production of 22,000 Curies of Cs-137 Per Day in 15 in. dia Higgins Continuous Ion Exchange Contactor. Resin: Duolite C-3.

The major quantities of the strontium and the rare earths appear in the sodium waste stream along with about 30% of the cesium. This cesium is eventually recovered since the sodium waste solution is recycled to the feed after being passed through a fixed bed of sulfonated polystyrene resin, Dowex 50, where the strontium and the rare earths are removed to prevent their buildup within the contactor.

A backwash section[11] is included in the contactor to remove fines formed by resin attrition. If they were not removed they would eventually cause plugging. The resin undergoes considerable radiation breakdown; consequently, appreciable quantities of the resin must be replaced in order to maintain 80% exchange capacity[12]. About 2 ft^3 of resin is removed daily at the spent resin takeoff and an equivalent amount of fresh resin is added at the resin make-up point. The backwash solution and the spent resin are filtered to remove the broken resin, and the effluent is returned to the feed.

3.0 EXPERIMENTAL RESULTS

Preliminary experiments were made in fixed ion exchange resin beds of Duolite C-3 from which the operating conditions were calculated for the 1.5-in.-dia continuous contactor. For optimum contactor performance calculations from the fixed bed data indicate that the feed flow rate must be <2000 ml/min, 0.3-3.0 M in Na, at a resin movement rate of 10 ml/min. The flow rate of the eluant, 3.0 M HCl, must be >55 ml/min for complete elution, and cesium-sodium separation will be maximum when the acid appearing in the sodium waste approximates 24 meq/min. Tracer studies with Sr-90 and Ce-144 indicated that strontium and the rare earths are sorbed as strongly as but eluted more easily than cesium.

Data obtained from the continuous contactor operation agree with those from the fixed bed. At a resin movement rate of 10 ml/min, synthetic ORNL waste containing Cs-137, Sr-90, and Ce-144 tracer at activity levels of 3×10^3 - 3×10^5 c/ml.min were processed at rates of 200-1600 ml/min (250-2000 gal/hr per square foot of resin). Activity in the waste was reduced to 0-50 c/ml.min, corresponding to decontamination factors of $>10^4$ for cesium and $>10^3$ for strontium and the rare earths. Complete tracer elution resulted at a 3.0 M HCl eluant flow rate >60 ml/min. When the rate of eluant acid appearing in the sodium waste was 30 meq/min the cesium product solution contained 15 ppm of sodium and 30% of the cesium followed the sodium waste. Increasing the rate resulted in a slight decrease in the sodium contamination of the cesium, but a much larger percentage of cesium appeared in the sodium waste. Decreasing the rate resulted in a decrease in the cesium that followed the sodium, but sodium contamination of cesium increased significantly.

3.1 Relation between Fixed Resin Bed Studies and Continuous Contactor Operation

Fixed bed studies were made in order to obtain distribution coefficients necessary for the establishment of initial sorption and elution conditions in continuous contactor equipment. Though conditions may be determined by trial and error, much time is saved if initial settings are approximately those required for optimum performance, with only minor adjustments being necessary during operation.

The distribution coefficient, as used in this report, is defined as the ratio of the amount of component on the resin to the amount in solution. It is calculated by the use of the equation

$$D.C. = \frac{V_s}{V_r}$$

where V_r is the resin volume and V_s is the volume of a tracer-bearing solution passed through the resin at 50% tracer breakthrough. For complete sorption in the continuous contactor, the ratio of feed solution to resin flow rate must be less than the distribution coefficient determined from the fixed-bed test in the same medium. For complete elution, the eluant-to-resin flow rate ratio must be greater than the distribution coefficient obtained from the fixed-bed study in the elution medium.

The eluant is diluted at the take-off point of the less strongly sorbed substance because of water slippage* in the contactor. The degree of dilution is not always known; consequently it is more convenient to measure the eluant flow rate, at that point, in terms of meq/min and convert to ml/min through the relation

$$ml/min = \frac{meq/min}{N}$$

Because of this, minor adjustments will be necessary during initial dilution stages of operation.

The separation factor is defined as the ratio of the distribution coefficient of one component to that of another in the same medium. It is a measure of the degree or ease of separation of the two components. For maximum separation of two components in the continuous contactor, the ratio of the rate of eluant appearance with the less strongly sorbed substance to the rate of resin movement should be set greater than, but as close as practical to, the value of the distribution coefficient determined for the substance.

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* In moving the resin under a hydrolyic surge, in the continuous contactor, the water travels at a faster rate than the resin. The additional water passing a point during resin movement is termed "slippage."

3.2 Fixed Resin Bed Studies

From fixed-bed studies it is concluded that for optimum operation of the 1.5-in.-dia continuous contactor, the following conditions are required, assuming a resin flow rate of 10 ml/min: For complete sorption of cesium, strontium, and the rare earths the feed flow rate must be < 2000 ml/min. This figure is low in that the cesium distribution coefficient of $\gg 200$ was determined after only 2% tracer breakthrough instead of at the 50% breakthrough point called for by theory. Sodium elution will be complete and sodium-cesium separation will be maximum when the rate of eluant at the sodium take-off is 24 meq/min (equivalent to 8 ml/min of 3.0 M HCl). Cesium elution will be complete when the eluant rate is >165 meq/min (55 ml/min of 3.0 M HCl).

In sorption studies cesium tracer showed a distribution coefficient to Duolite C-3 resin of $\gg 200$ from a 0.35 M NaOH--0.3 $NaNO_3$--0.1 M $NaAlO_2$ solution (Fig. 3.1). The unusual affinity of this resin for cesium was maintained in alkaline solutions of high salt content, 3 M $NaNO_3$ or 1 M NH_4NO_3, where the distribution coefficients were >100 and >200, respectively (Fig. 3.2). This shows that rather large variations in feed concentration can be handled without decreasing the contactor efficiency. With macro quantities of cesium, 1 g per liter of solution, the loading capacity of Duolite C-3 was determined to be 45 g cesium per liter of resin (Fig. 3.3). With Sr-90 or Ce-144 tracer, the sorption effluent had activities of background level, showing complete sorption of these fission products. The distribution coefficients were estimated to be of the same order of magnitude as that of cesium.

In breakthrough studies, cesium and sodium showed distribution coefficients to the resin from 3.0 M HCl of 5.5 and 0.8, respectively, which corresponds to a separation factor of 6.8 (Fig. 3.4). It was expected that elution of strontium and cerium would be more difficult than elution of cesium, which is the case with Dowex-50. However, because of the great affinity of Duolite C-3 for cesium, elution was more rapid for strontium and the rare earths (Fig. 3.5).

3.3 Continuous Contactor Operational Studies

Synthetic ORNL waste containing Cs-137, Sr-90, and Ce-144 tracer was processed in the 1.5-in.-dia continuous ion exchange contactor at feed rates of 200-1600 ml/min (250-2000 gal/hr per square foot of resin) and a resin movement rate of 10 ml/min. At 3.0 M HCl eluant flow rates >60 ml/min, activity in the feed waste was 0-50 c/ml.min, corresponding to decontamination factors of $>10^4$ for cesium and $>10^3$ for strontium and the rare earths. However, at an eluant flow rate of <60 ml/min, the feed waste activity increased, indicating incomplete elution.

Approximately 15 ppm of sodium appeared in the cesium product and 30% of the cesium appeared in the sodium waste when the acid flow rate to the sodium waste was 30 meq/min (10 ml/min of 3.0 M HCl). An increase in the acid rate produced only a slight decrease in the sodium contamination of cesium but a large increase in the percentage of cesium appearing with the sodium.

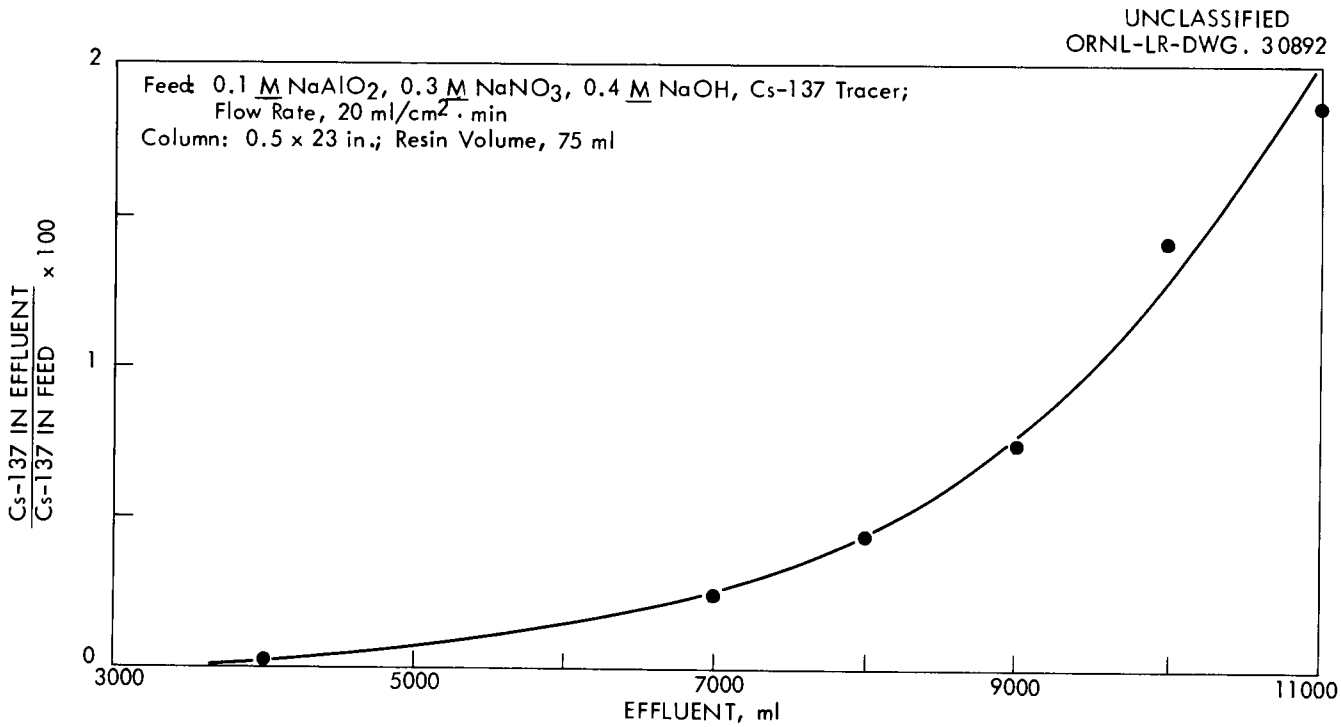

Fig. 3.1. Sorption of Cs-137 Tracer from Synthetic Alkaline Waste of Low Salt Content Onto Duolite C-3 Resin.

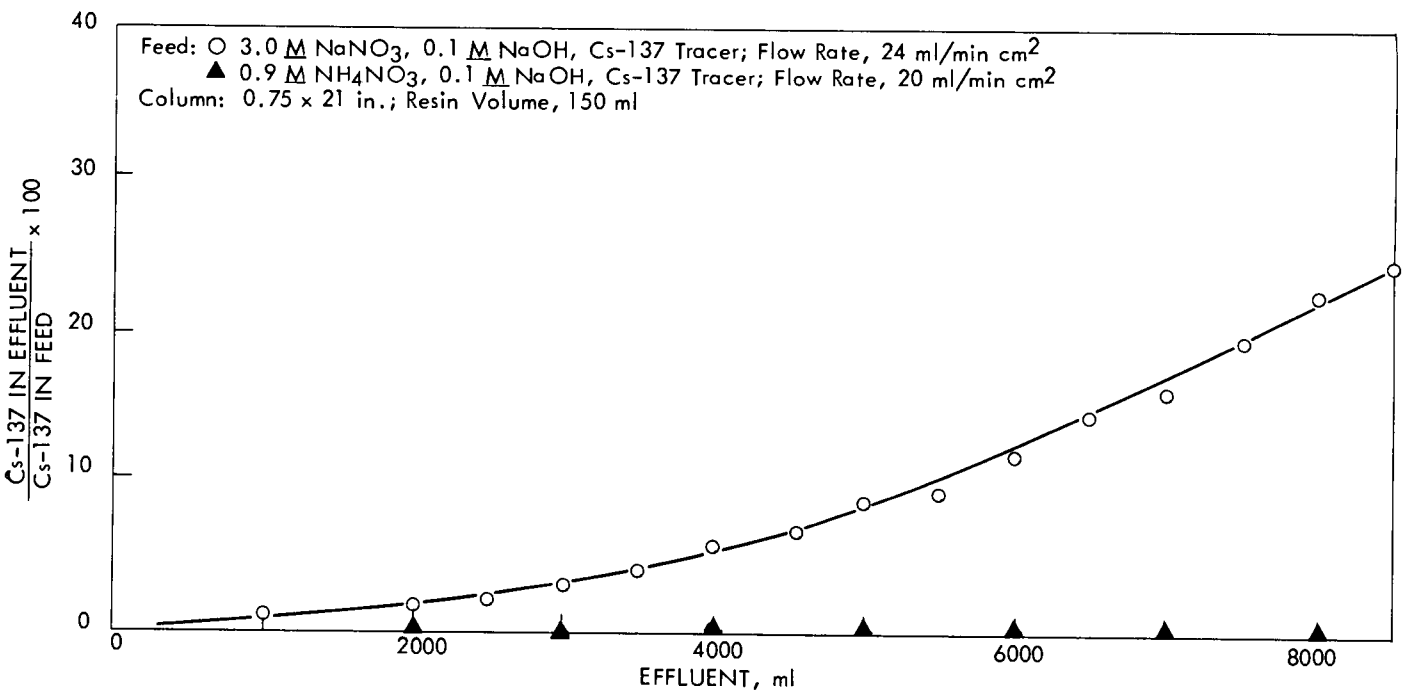

Fig. 3.2. Sorption of Cs-137 Tracer from Synthetic Alkaline Waste of High Salt Content Onto Duolite C-3 Resin.

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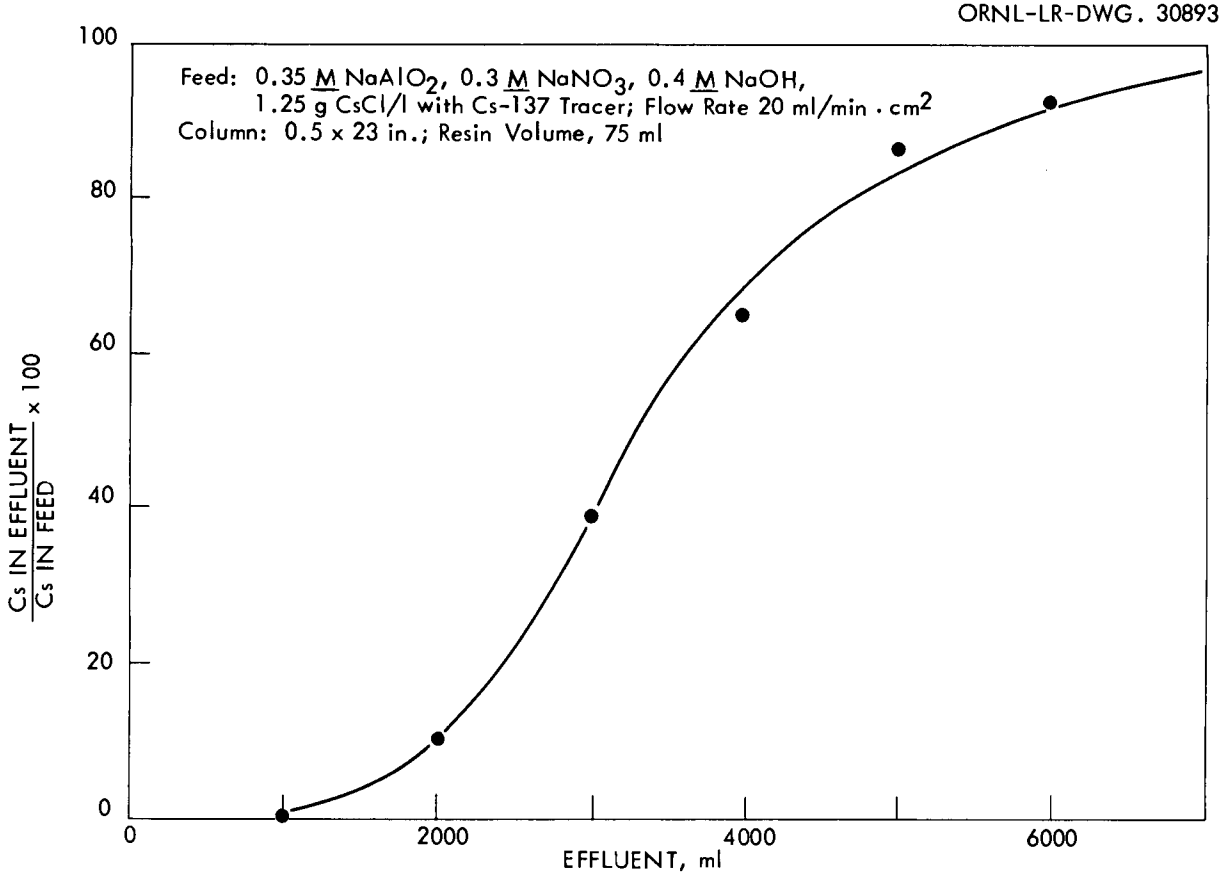

Fig. 3.3. Sorption of Macro Quantities of Cesium from Synthetic Alkaline Waste Onto Duolite C-3 Resin.

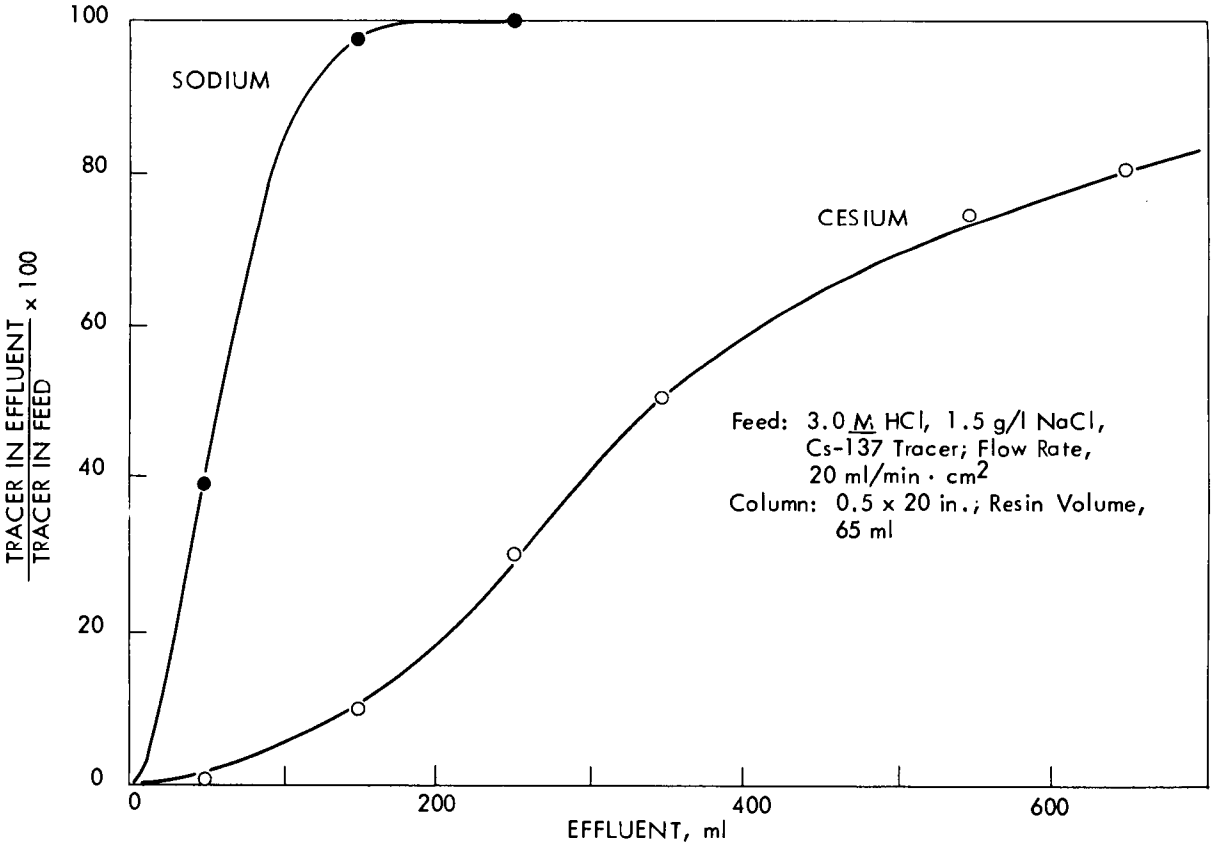

Fig. 3.4. Breakthrough of Sodium and Cesium from Duolite C-3 Resin in 3.0 M HCl.

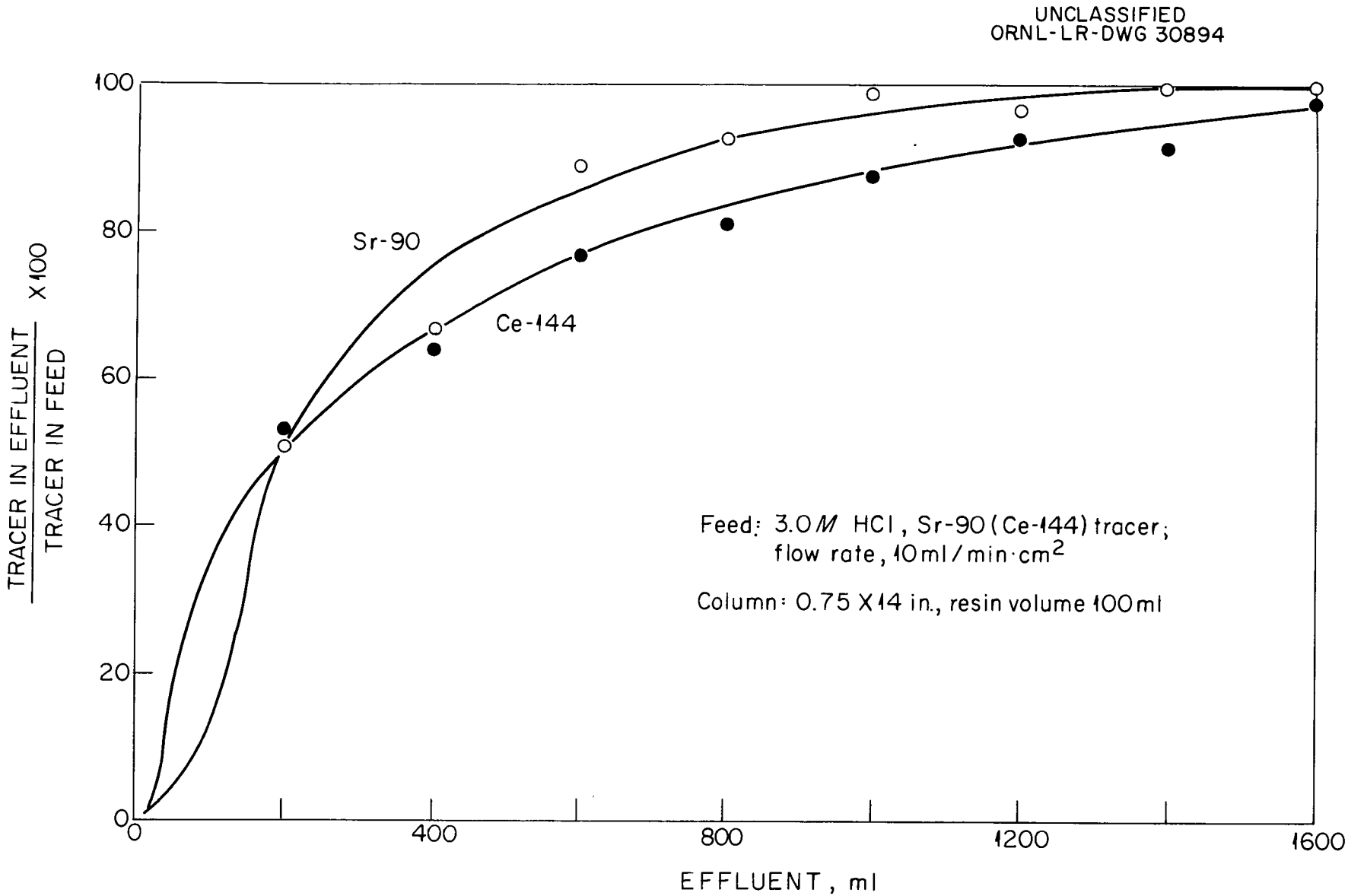

Fig. 3.5. Breakthrough of Sr-90 and Ce-144 Tracer from Duolite C-3 Resin in 3.0 M HCl.

On the other hand, when the acid rate was decreased the percentage of cesium in the sodium waste decreased sharply, but the sodium contamination of the cesium also increased rapidly.

On recycle of sodium waste to the feed no extra burden was noted on the contactor.

The experiments were made in a 1.5-in.-dia continuous contactor (Fig. 3.6) with 3-ft sorption and elution sections and a 2-ft scrub section. Initial settings were approximately those indicated as required by the fixed-bed studies. However, instead of making the necessary adjustments to obtain optimum contactor performance, the flow rate conditions were varied rather widely in order to obtain data to support or disprove the stated relations between fixed-bed studies and continuous contactor operation. Low levels of activity were used since the primary objectives of the study were to demonstrate high processing rate and effective cesium-sodium separation. Consequently, maximum decontamination factors were not obtained.

Aqueous feed rates of simulated ORNL waste with Cs-137 tracer (3×10^3 --3×10^5 γ c/ml.min) were varied from 200 to 1600 ml/min (250-2000 gal/hr per square foot of resin). A centrifugal pump was used for feeding the contactor since precisely metered flow was not required. Activity in the feed effluent ranged from 0 to 215 γ c/ml.min (Table 3.1). As discussed below, the higher activity resulted when the flow rate of the eluant, 3.0 M HCl, dropped below about 60 ml/min. The estimated cesium decontamination factor was $>10^4$.

The eluant flow rate was varied from 50 to 105 ml/min. In general, the feed waste activity was >50 γ c/ml.min when the eluant flow rate was below about 60 ml/min and <50 γ c/ml.min at flow rates of >60 ml/min (Table 3.1). This is an indication that at an eluant flow rate of <60 ml/min elution of cesium was not complete and is in good agreement with the value calculated from the fixed bed studies (55 ml/min minimum eluant flow rate for complete elution).

The maximum cesium-sodium separation could not be determined since cesium was present in trace amounts while sodium was present in macro quantities. However, an indication of the operational performance in the 2-ft sodium elution section was obtained on plotting cesium in the sodium waste and sodium in the cesium product as a function of the rate of appearance of eluant acid in the sodium waste (Fig. 3.7). The sodium contamination of cesium increased slightly from 5 to 15 ppm as the waste-acid rate decreased from about 120 to 30 meq/min. Decreasing the acid rate to about 1 meq/min resulted in a rapid increase of cesium contamination to >200 ppm sodium. This again is in agreement with the value >24 meq/min (8.0 ml/min of 3.0 M HCl) of acid appearing in the sodium waste as calculated from fixed-bed studies for the complete elution of sodium. Cesium in the sodium waste, on the other hand, increased in a hyperbolic manner from about 1% to 70% as the acid rate was increased from 1 meq/min to 120 meq/min. Thus, at 30 meq/min

Table 3.1 Average Operating Conditions and Performance of the 1.5-in.-dia Higgins Continuous Ion Exchange Contactor in Removing Cesium from Alkaline Waste

Sorption and cesium elution sections were each 3 ft long; sodium elution section was 2 ft long; Duolite C-3 resin movement was at a rate of 10 ml/min; feed composition was 0.3 M NaOH with Cs-137 tracer levels of 3×10^3 to 3×10^5 γ c/ml.min; eluant was 3.0 M HCl.

			Sodium Waste		Cesium Product		
Feed Flow Rate, ml/min	Feed Waste Activity, c/ml.min	Eluant Flow Rate, ml/min	Acid Flow Rate, meq/min	Cesium Content, % of total	Acid Flow Rate, meq/min	Cesium, % of total	Sodium, ppm
192	17	49	71	49	66	51	7
183	56	71	70	56	125	44	7
377	0	86	44	42	214	58	12
408	12	104	109	72	194	28	7
397	25	105	88	58	229	42	9
511	43	104	76	48	238	52	7
612	63	105	94	48	208	52	5
609	51	72	53	49	170	51	13
603	34	84	73	53	177	47	7
768	37	85	40	35	202	65	15
725	83	61	3	5	169	95	120
913	129	55	1	4	159	96	161
994	122	55	1	5	157	95	215
942	113	51	13	20	131	80	15
1180	81	53	28	28	111	72	18
1370	37	70	9	11	161	89	31
1510	57	56	<1	1	148	99	208
1520	19	60	8	10	157	90	29

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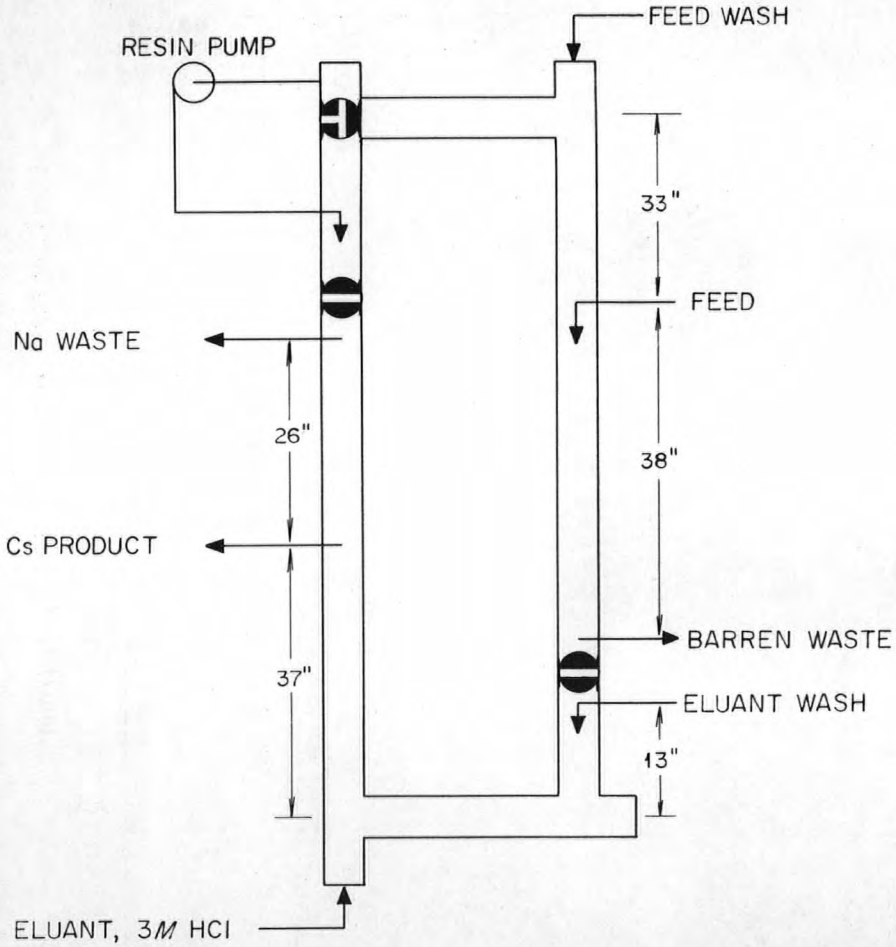

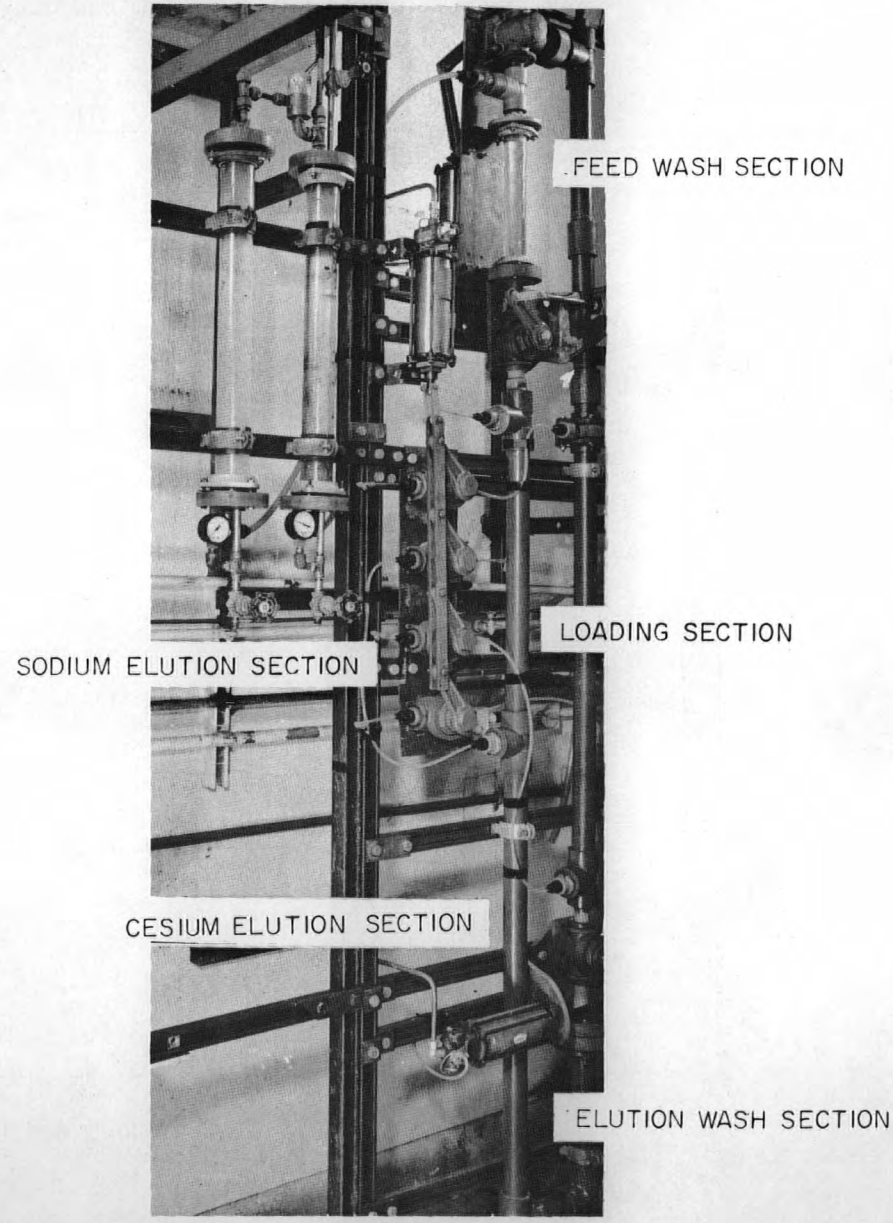

Fig. 3.6. Higgins Continuous Ion Exchange Contactor, 1.5 in. dia.

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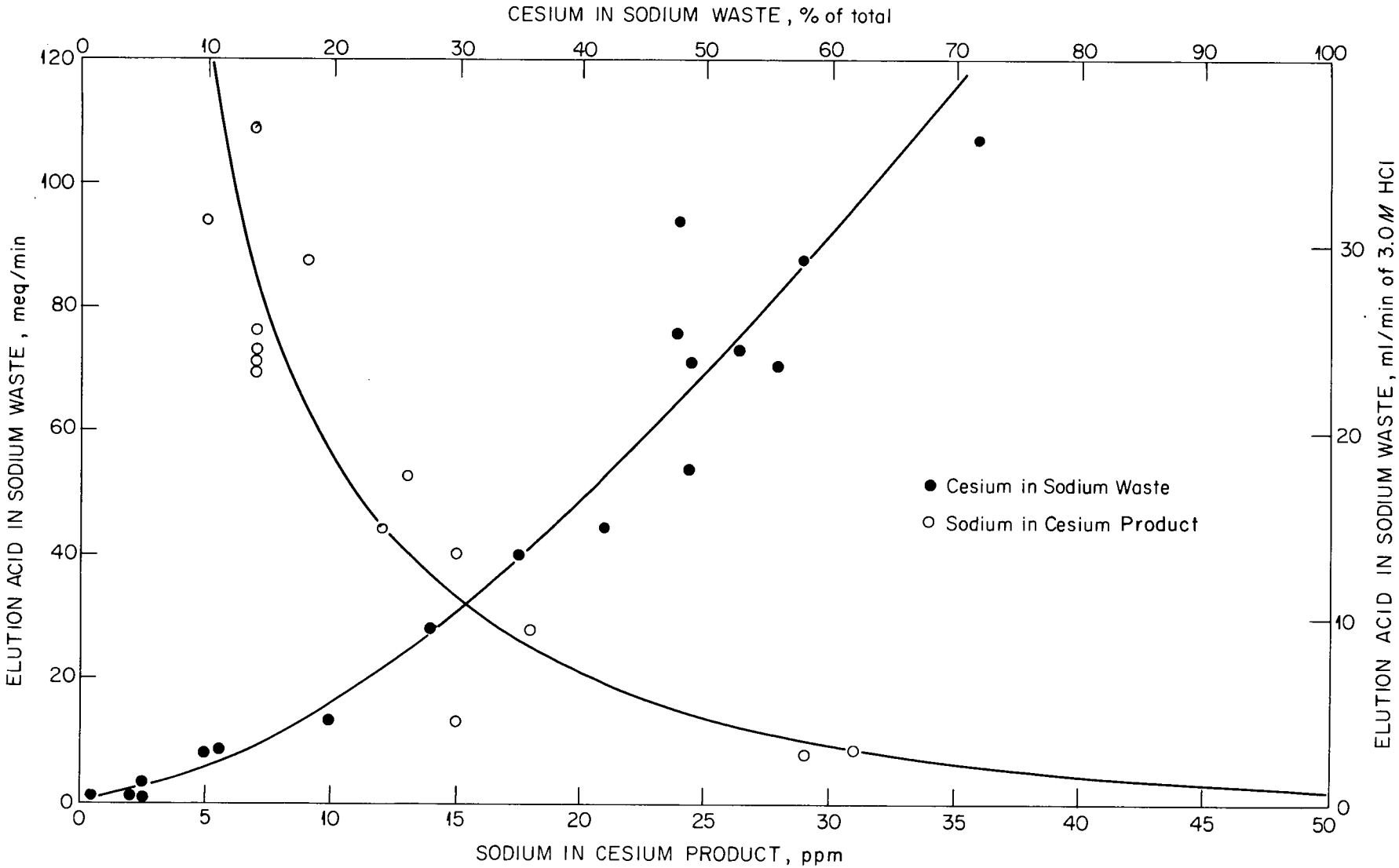

Fig. 3.7. Sodium in Cesium Product and Cesium in Sodium Waste as a Function of Acid Appearing in the Sodium Waste. Resin Rate, 10 ml/minute; Length of Scrub Section, 2 ft.

(10 ml/min of 3.0 M HCl) of acid appearing in the sodium waste, 30% of the cesium was recycled to the feed while the remaining 70% was collected as product with sodium contamination of 15 ppm.

In a run on synthetic sodium aluminate waste, 0.1 M $NaAlO_2$--0.8 M NaOH containing Cs-137 tracer, dilute caustic wash solutions were required in order to prevent precipitation of aluminum hydroxide in the column. Decontamination factors were $>10^3$ at tracer activities of 10^3 - 10^4 γ c/ml·min (Table 6.4). The dilute sodium hydroxide use in the elution wash converted the hydrogen-form resin to the sodium form before it was contacted with the sodium aluminate. A momentary stoppage of this wash resulted in precipitation of aluminum hydroxide, which plugged the column. Dilute hydroxide was used as the feed wash to prevent precipitation as a result of lower hydroxide concentration. Aluminum in the cesium product was <8 ppm.

Two short runs were made with Sr-90 and Ce-144 tracer in 0.3 M NaOH as respective group representatives. High decontamination factors were difficult to demonstrate because of the low tracer solubility in the alkaline solution. However, it was estimated that sorption was approximately equivalent to that of cesium (Tables 6.2 and 6.3). The major quantity of strontium and cesium followed the sodium. In order to prevent unlimited buildup of strontium and the rare earths in the contactor, it is recommended that the sodium waste be passed through a fixed bed of sulfonated polystyrene cation resin, prior to recycle, to selectively remove these elements.

Duolite C-3 is irregular rather than in bead form, and a high attrition rate was expected in the contactor. After 2 weeks of operation, the pressure drop in the loading section of the contactor increased to the point of virtual plugging. After the resin was removed from the contactor, washed free of fines, and then returned, operation was continued for about 6 weeks without further trouble. This observation parallels other studies in which attrition is high on a fresh resin but drops off with time. However, because of the irregular form of the resin, attrition is expected to be greater than with the bead-form polystyrene resins.

Chemical as well as physical attrition was of interest in the elution section because of contaminants which might be added to the cesium product. Four liters of 3 M hydrochloric acid cesium-strip solution was evaporated to 20 ml. The resulting solution was tinged brown and contained suspended organic material. Qualitatively this did not appear to be a significant contaminant for a cesium radiation source. Further investigation is required in order to determine whether or not the organic matter present could cause gas pressure in a sealed bomb under irradiation and if it can be satisfactorily removed by filtration.

Detailed run data for the continuous contactor may be found in the tables in the appendix.

4.0 PROCESS COST ESTIMATE

An estimate was prepared to determine the cost of Cs-137 removal and recovery from alkaline waste solutions at a large plant (Fig. 2.1). The inherent features of the continuous ion exchange method include the practicability for remote control operation and maintenance contributing to low cost. The sensitivity of the chemical system to radiation and the ease with which damage can be corrected will partly determine the cost.

It was assumed that the contactor feed is neutralized tank storage waste, in agreement with current waste-treatment practices. The major cost-saving feature of ion exchange removal of cesium from dilute alkaline waste is a combination of the high affinity and selectively of the resin for cesium and the very high processing rate possible in the continuous contactor. A second cost-saving feature is the elution of the final product with hydrochloric acid, since current practice is to prepare cesium sources as a solid chloride. The hydrochloric acid is evaporated from the final product, condensed, and reused. An appreciable quantity of makeup resin is required because of radiation breakdown. For 22,000 curies of Cs-137 production per day, about 2 ft^3 of Duolite C-3 resin must be replaced to maintain 80% exchange capacity.[12]

The mechanical equipment that must be operated and maintained by remote control are the ion exchange contactor, a hydrochloric acid still, and the cesium chloride drying and packaging equipment. It is suggested that the contactor be constructed entirely of Hastelloy-C because of the use of chloride. Since the contactor is not very large, this cost is not prohibitive. Resin may easily be loaded and unloaded remotely, while many of the pumps and valves may be located in the operating gallery for easy access. The evaporator must be resistant to boiling hydrochloric acid and should be constructed of Hastelloy-C. Details of the mechanical handling accessories are not specified; however, this type of equipment, and that permitting remote welding of the canned product, is being used at the ORNL isotope production area[13].

The cost estimate assumes a plant capacity of 8×10^6 curies per year in a two-cell building. The first cell contains the ion exchange contactor and evaporator while the product drying and packaging equipment is located in the second. The cells are 8 by 8 by 25 ft high with 3-ft-thick concrete walls. Each has one 4-by 8-ft sliding vertical door on rails and an overhead cover handled by a monorail crane. Radiation-proof viewing windows and mechanical manipulators are included in the cost estimate.

It is assumed that the cesium plant is located adjacent to a large neutralized-waste storage farm. Supernatant from the alkaline nitrate waste would be pumped directly to the ion exchange column. The barren waste from the contactor would be pumped back to the tank farm for further processing or disposal to the environment.

4.1 Total Plant Investment

Complete building		$ 270,400
Cell 1		
Storage tank, 100 gal	$ 2,500	
Contactor and four pumps	75,000	
HCl still	12,500	
Reflux tower	7,500	
Condenser	3,500	
Holdup tank	2,500	
Instrumentation	25,000	
Piping	36,500	
Equipment cost of Cell 1	$ 165,000	
Installation, 40% of equipment cost	66,000	
Physical cost of Cell 1	231,000	
Overhead, 50% of physical cost	115,500	
Contingency, 10% of physical cost	23,100	
Total cost of Cell 1	369,600	369,600
Cell 2		
Dryer	$ 10,000	
Hydraulic press	10,000	
Remote welding	12,500	
Carrier	20,000	
Instrumentation	10,000	
Equipment cost of Cell 2	$ 62,500	
Installation, 40% of equipment cost	25,000	
Physical cost of Cell 2	$ 87,500	
Overhead, 40% of physical cost	43,750	
Contingency, 10% of physical cost	8,750	
Total cost of Cell 2	$ 140,000	140,000
Total Plant investment		$ 780,000

4.2 Daily Operating Cost

Depreciation, cost/day, based on 6-2/3 year amortization of total plant investment and 300 days/year operation		$ 390.00
Operating cost per day		
Labor, 13 men at $25/day	$ 325.00	
Resin, 2.1 ft^3/day at $20/$ft^3$	42.00	
Sodium chloride, 210 lb/day at 0.75 cent/lb	1.58	
Hydrochloric acid, 630 lb/day at 1.4 cents/lb	8.82	
Labor and materials cost	377.40	
Utilities, 10% of labor and materials	37.74	
Labor, materials and utilities cost	415.14	
Overhead, 100% of labor, materials, and utilities cost	415.14	
Operating cost per day	830.28	830.28
Total daily operating cost		$ 1220.28

Cesium cost per curie, based on 22,000 curies/day — 6 cents/curie

Unevaluated credit: removal of long-lived isotopes from waste.

5.0 REFERENCES

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6.0 APPENDIX

6.1 Features of the Higgins Continuous Ion Exchange Contactor

The Higgins contactor is a continuous-countercurrent solid-liquid contacting device for efficient mass transfer. Arranged in a loop form, the resin is separated into sections by quick-opening wide-port valves. The various ion exchange steps (i.e., sorption, washing of the feed solution from the resin void volumes, selective elution, and resin regeneration) occur simultaneously in consecutive sections of the contactor. During solution flow, usually 2-10 min, the valves separating the sections are closed and the resin bed is stationary. At specified time intervals the solution flow is stopped and the valves are opened. A sudden hydraulic surge across the resin loop slides the resin bed a predetermined short distance from one section to the next in a direction opposite to that of the solution flow. The valves again close and solution flow is continued. The valves are of the "volume displacement" rather than the "volume reduction" type and thus do not tend to jam or crush the resin on closing.

Attractive features of this contacting method are: (1) all flows of both solution and resin may be metered independently of each other in rate or applied pressure, (2) high-density solutions may be processed that would normally float the resin because the resin is confined in a limited volume, (3) resin is moved gently with a hydraulic thrust and is not contacted with a mechanical pump, and (4) unclarified solution may be handled without plugging of the contactor.

6.2 Details of Experimental Data

The following tables contain the sorption and elution data and the analyses for the various studies made on the 1.5-in.-dia continuous ion exchange contactor. Resin movement was 1 in. every 3 min, a rate of 10 ml/min. Except where otherwise noted, the feed composition was 0.3 M NaOH with respective tracer added, and the feed and elution washes were water. In all studies 3.0 M HCl was the eluant.

Table 6.1 Run Data with Cesium-137 Tracer in 0.3 M NaOH

		Flow Rates, ml/min							Analysis						
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution Waste		Product		
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+, ppm
8/7/57	1:00 pm	Start													
	2:00	133	36	163	25	23	33	18	35,000	134	41	---	36	1.85	2790
	3:00	166	35	220	21	26	42	17		64	150	---	163	2.40	55
	4:00	250	28	253	38	36	58	18		102	8,790	---	137	2.40	10
8/8/57	8:20 am	Start													
	9:20	183	21	237	53	33	63	30		8	24,800	---	18	2.62	7
	10:20	233	28	191	45	33	53	30		39	49,900	---	1,010	2.32	7
	11:20	166	25	216	48	33	50	25		13	27,000	---	15,100	2.60	7
	12:20 pm	216	33	190	46	33	46	30		12	50,000	---	29,700	2.62	7
	1:20	183	33	210	50	33	55	30		0	48,500	---	35,200	2.44	7
	2:20	200	33	263	50	31	51	30		22	42,300	---	40,600	2.56	7
	3:20	200	25	216	50	31	51	33		26	83,700	---	61,800	2.72	7
8/9/57	8:50 am	Start													
	9:50	200	31	241	66	26	50	50		31	86,700	1.69	55,700	2.68	7
	10:50	200	30	230	73	25	58	53		12	38,800	1.07	57,600	2.82	7
	11:50	200	35	238	90	31	61	56		15	54,900	1.36	53,000	2.76	6
	12:50 pm	133	20	170	55	20	35	35		267	98,900	1.37	58,200	2.72	6
8/13/57															
	12:30 pm	Start													
	1:30	333	25	383	87	33	53	75	259,000	0	43,400	0.91	44,800	---	22
	2:30	366	36	400	75	33	30	78		0	40,400	0.72	40,300	---	22
	3:30	433	30	425	95	33	48	93		0	28,500	0.58	39,200	---	22
8/15/57	8:25 am	Start													
	9:25	433	38	433	93	33	53	90		23	122,000	1.01	46,300	2.60	18
	10:25	383	38	475	115	33	55	90		0	261,000	1.90	35,900	2.60	20
	10:40														
	11:40	414	36	466	113	36	66	90	134,000	25	154,000	1.18	63,700	2.63	20
	12:40 pm	400	33	463	103	33	50	90		21	209,000	1.56	68,200	2.73	8
	1:40	400	38	466	103	33	50	90		41	277,000	1.54	92,000	2.67	7
	2:40	400	36	466	110	33	50	90		0	176,000	0.98	109,000	2.68	8
	3:40	366	33	466	95	33	50	90		39	272,000	1.16	103,000	2.59	8

Table 6.1 (continued)

		Flow Rates ml/min							Analysis						
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution	Waste	Product		
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+, ppm
8/19/57	9:30 am	Start													
	10:30	683	30	716	100	33	70	91	43,000	42	186,000	1.21	165,000	2.72	7
	11:30	633	30	670	100	33	55	90		62	171,000	1.02	144,000	2.66	7
	12:30 pm	616	33	716	108	33	55	90		23	309,000	0.84	168,000	2.68	7
	1:30	616	33	716	86	33	45	90		57	516,000	1.22	165,000	2.76	7
	2:30	600	33	700	95	33	58	93		44	163,000	0.71	196,000	2.70	6
	3:30	518	51	585	133	37	62	100		31	241,000	0.74	210,000	2.70	6
8/20/57	12:10	Start													
	1:10	516	55	666	100	55	85	91	23,600	66	198,000	1.64	114,000	2.59	5
	2:10	650	51	733	103	50	78	91		58	193,000	1.43	124,000	2.38	6
	3:10	680	60	783	113	50	68	91		61	112,000	1.48	90,000	2.26	5
	4:10	625	62	729	105	56	62	97		67	60,400	0.61	80,600	2.19	5
8/22/57	9:50 am	Start													
	10:50	633	36	666	58	41	40	75	8,190	85	101,000	1.05	102,000	2.32	13
	11:50	650	62	800	93	33	73	78		66	97,800	1.50	61,900	2.51	15
	12:50 pm	650	51	725	63	33	53	78		71	54,800	1.09	46,500	2.43	13
	1:50	566	58	650	75	45	50	78		68	65,300	0.60	88,300	2.40	14
	2:50	545	49	662	72	49	65	81		65	132,000	1.43	63,300	2.49	15
8/23/57	8:40 am	Start													
	9:40	616	46	616	91	43	66	78	3,290	38	144,000	1.58	59,600	2.75	7
	10:40	600	53	666	78	50	65	78		40	94,000	1.43	55,100	2.45	6
	11:40	600	50	733	83	43	55	78		14	61,500	1.42	52,000	2.33	6
	12:40 pm	600	53	725	83	46	56	76		30	7,630	0.23	42,900	2.15	6
	1:40	600	50	666	83	50	56	78		49	48,900	1.45	43,700	2.33	13
	2:20														
	3:20	600	38	625	83	41	66	78	3,410	19	56,600	1.59	49,000	2.35	6
	4:20	559	53	731	78	48	68	78		61	51,000	1.56	42,900	2.50	6

Table 6.1 (continued)

		Flow Rates, ml/min							Analysis						
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution	Waste	Product		
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+, ppm
9/10/57	10:20 am	Start													
	11:20	---	46	516	108	50	96	96	7,120	3	41,500	1.25	11,000	3.16	49
	12:20 pm	963	36	800	83	33	71	80		32	76,800	1.64	22,100	2.70	11
	1:20	438	33	716	84	41	50	83		23	32,600	0.81	37,600	2.60	7
	2:20	816	41	866	81	41	50	80		31	4,760	0.13	32,500	2.22	6
	3:20	850	51	925	83	53	66	80		29	2,660	0.02	25,500	2.64	9
9/12/57	10:10 am	Start													
	11:10	833	33	966	61	36	41	80	7,840	131	8,640	0.34	19,100	2.31	134
	12:10 pm	734	41	800	61	46	33	80		68	104	0.16	41,500	2.30	235
	1:10	500	50	600	63	53	33	80		84	3,730	0.01	26,500	1.92	73
	2:10	833	41	949	60	46	25	80		66	207	0.03	56,000	2.40	390
9/13/57	9:40 am	Start													
	10:40	1066	41	1,176	56	38	41	80	7,500	113	18,000	0.04	23,400	2.04	220
	11:40	841	41	883	55	45	33	81		118	1,800	neut.	37,300	1.95	320
	12:40 pm	833	45	900	55	41	50	80		154	29,000	0.02	51,200	2.33	44
	1:50														
	2:50	1,000	33	1,133	56	33	33	75	5,510	129	1,610	0.02	64,800	1.86	590
	3:50	1,000	53	1,116	65	50	33	81		154	9,530	0.06	41,500	1.92	37
9/16/57	12:20 pm														
	1:20	983	36	1,050	41	35	41	36		82	2,750	0.07	35,100	1.88	19
9/17/57	9:20 am	Start													
	10:20	801	36	1,083	41	36	58	40	6,400	118	24,500	0.38	34,800	2.21	19
	11:20	858	53	933	58	53	33	75		125	18,600	0.19	66,400	2.33	14
	12:20 pm	1,166	52	1,385	52	58	41	70		95	13,300	0.29	60,300	2.28	12
	1:45														
	2:45	916	33	1,083	46	33	30	58	7,540	114	663	0.02	30,000	1.84	44
	3:45	1,063	36	1,166	36	36	33	58		96	10,600	0.03	30,300	1.96	40
9/18/57	10:20 am														
	11:20	1,461	34	1,461	76	30	76	69		32	52,600	0.91	64,000	2.67	9

Table 6.1 (continued)

		Flow Rates, ml/min							Analysis						
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution	Waste	Product		
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+, ppm
9/18/57	12:40 pm	Start													
	1:40	1,300	36	1,283	81	38	46	75	6,200	49	2,260	0.07	52,900	2.55	10
	2:40	1,446	46	1,566	60	51	41	70		26	19,300	0.33	53,500	2.21	7
9/19/57	9:10 am	Start													
	10:10	1,400	36	1,566	60	40	25	75	5,480	51	1,590	0.04	87,900	2.64	4
	11:10	1,616	46	1,716	53	50	30	70		65	346	0.01	74,700	1.76	235
	1:00 pm														
	2:00	1,366	41	1,491	60	41	40	70	5,730	5	313	0.02	87,100	2.16	35
	3:00	1,673	45	1,725	61	45	43	70		32	37,900	0.36	106,000	2.60	2

Table 6.2 Run Data with Strontium-90 Tracer in 0.3 M NaOH

		Flow Rates, ml/min							Analysis						
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution	Waste	Product		
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+, ppm
9/23/57	12:30 pm	Start													
	1:30	200	50	316	60	50	66	66	18,200	8	2,070	0.68	2,300	2.02	8
	2:30	200	36	266	56	35	30	70		14	774	0.22	4,600	2.16	8
	3:30	213	46	266	60	48	33	70		10	1,680	0.25	6,400	2.04	8
9/24/57	9:00 am	Start													
	10:00	216	33	250	56	33	33	70		0	2,360	0.70	5,700	2.72	10
	11:00	233	33	258	58	33	45	70		0	345	0.06	6,170	2.56	12
	12:00 pm	236	45	260	61	50	50	70		8	4,400	0.51	5,370	2.24	10
	1:00	216	45	266	60	50	41	70		20	2,310	0.35	4,560	2.16	11
	2:00	191	45	233	61	38	33	70		14	4,610	0.30	5,570	2.38	12
	3:00	183	40	266	58	45	33	70		10	3,070	0.23	4,070	2.42	12
	4:00	200	41	266	58	46	33	70		18	4,740	0.13	3,780	2.30	10
9/25/57	8:40 am	216	33	266	55	36	36	70		0	5,760	0.46	3,670	2.62	15
	10:40	203	36	266	58	46	36	70		14	9,570	0.55	2,920	2.58	10
	11:40	236	36	266	58	36	36	70		0	6,960	0.54	2,360	2.52	10
	12:40 pm	203	36	250	55	40	36	70		16	3,140	0.37	2,080	2.26	11
	1:40	203	50	250	60	45	36	70		0	1,390	0.25	1,790	2.04	12
	2:40	220	36	250	56	40	36	70		8	6,550	0.21	1,540	1.90	12

Table 6.3 Run Data with Cerium-144 Tracer in 0.3 M NaOH

		Flow Rates, ml/min							Analysis						
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution	Waste	Product		
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+, ppm
9/27/57	8:10 am	Start													
	9:10	233	33	275	53	33	36	70	19,200	1,380	71	0.22	192	2.64	217
	10:10	136	35	233	60	33	36	70		202	14	0.04	164	2.27	27
	11:10	203	40	266	61	41	36	70		25	112	0.95	104	2.20	9
	12:10 pm	200	41	250	60	43	36	70		16	1,220	0.71	83	2.20	9
	1:10	233	46	266	60	40	33	70		0	591	0.61	73	1.87	10
	2:10	200	36	266	63	41	33	70		4	521	0.63	58	1.84	9
	3:10	200	36	266	55	50	41	70		0	296	0.41	71	1.74	9
9/31/57	12:45 pm	Start													
	1:45	166	41	250	53	30	36	70	4,490	27	237	0.40	230	1.64	8
	2:45	200	40	266	56	40	36	70		39	348	0.66	251	1.70	8
	3:45	200	35	266	60	38	36	70		43	163	0.55	148	1.64	7
10/1/57	8:30 am	Start													
	9:30	200	50	250	66	45	36	70		34	168	0.41	350	1.68	7
	10:30	200	41	233	60	41	33	70		73	1,300	0.02	265	1.46	85
	11:30	200	36	250	58	38	36	70		49	1,330	0.03	266	1.78	28
	12:30 pm	200	38	250	53	40	38	70		71	2,880	0.50	639	2.74	9
	1:30	200	38	250	56	36	36	70		26	1,380	0.09	817	2.68	13
	2:30	183	33	250	60	33	36	70		23	730	0.03	461	2.42	30
	3:30	200	36	250	60	40	36	70		48	663	0.24	335	2.14	11
	4:30	200	36	256	64	42	34	74		35	381	0.25	375	2.22	12
10/2/57	12:00 pm														
	1:00	200	36	266	53	33	36	70	3,650	57	560	0.22	400	2.68	71
	2:00	200	33	250	63	33	36	70		6	728	0.45	268	2.56	12
	3:00	200	46	266	53	41	36	70		90	392	0.42	219	2.32	7
	4:00	200	46	250	63	41	36	70		119	329	0.34	203	2.08	6
10/3/57	9:00 am														
	10:00	216	33	266	53	33	36	70	3,650	35	187	0.21	199	2.08	9
	11:00	216	36	266	58	36	36	70		47	725	0.38	264	2.02	8
	12:00 pm	216	38	266	58	38	36	70		22	551	0.31	232	1.90	8
	1:00	216	41	266	58	41	33	70		39	354	0.40	263	1.84	8
	2:00	200	41	266	58	36	33	70		3	491	0.41	483	3.04	8
	3:00	200	45	266	58	38	36	71		14	593	0.49	243	2.28	6
	4:00	200	38	266	58	33	36	70		15	534	0.54	279	2.78	7

Table 6.4 Run Data with Cesium-137 tracer in 0.1 M $NaAlO_2$ and 0.8 M NaOH

Sorption and elution washes were 0.5 M NaOH

		Flow Rates, ml/min							Analysis							
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution	Waste	Product			
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+, ppm	$Al^{=}$ ppm
8/26/57	12:15 pm	Start														
	1:15	433	48	550	86	50	61	81	2,830	47	31,900	1.28	34,900	2.37	7	--
	2:15	600	58	750	99	51	71	81		52	32,100	1.08	24,200	2.29	12	--
	3:15	666	55	750	86	50	65	80		0	26,700	0.73	41,400	2.69	13	--
	4:15	444	55	531	102	53	73	86		27	29,700	1.29	24,600	2.64	6	--
8/27/57	8:30 am	Start														
	9:30	350	43	450	95	46	76	80		44	46,100	1.81	23,000	2.80	5	--
8/30/57	12:40 pm															
	1:40	366	66	466	65	43	66	55	3,410	58	1,750	0.84	6,010	2.22	14	--
	2:40	400	61	466	85	56	63	80		55	1,350	0.52	5,060	2.28	15	--
	3:40	400	38	466	83	31	63	83		27	1,740	0.16	4,620	2.30	11	--
9/3/57	8:50 am	Start														
	9:50	366	33	383	73	33	33	83		15	3,660	0.46	3,340	---	33	7
	10:50	383	33	450	78	35	36	83		0	20,400	0.75	3,780	---	22	6
	11:50	383	35	463	80	35	46	80		15	20.300	0.77	4,750	---	18	6
	12:50 pm	383	33	450	73	33	43	73		12	12,900	0.76	5,470	---	17	8
	3:00															
	4:00	400	36	436	72	32	42	72	3,860	9	10,800	0.84	4,260	---	17	5
9/4/57	8:30 am	Start														
	9:30	431	36	466	78	40	58	73		14	16,500	1.05	5,480	2.14	62	6
	10:30	408	41	491	81	41	55	73		3	16,900	0.91	7,680	2.21	11	6
	11:30	405	50	491	80	38	63	76		0	16,300	1.14	8,060	2.18	10	6
	12:30 pm	391	41	466	76	36	50	75		0	23,700	1.04	9,410	2.26	8	6
	1:30	400	41	466	75	40	50	75		24	31,500	1.38	11,400	2.76	9	6
	2:30	400	41	466	83	36	50	75		46	35,000	1.28	10,800	2.69	6	6
9/5/57	11:20 am	Start														
	12:20 pm	400	50	483	65	45	63	63	7,250	13	25,300	1.22	10,900	2.55	10	8
	1:20	383	46	463	78	50	53	63		17	11,100	0.69	8,050	2.40	8	7
	2:20	433	55	463	81	46	60	76		65	26,100	1.25	7,390	2.60	9	8
	3:20	410	40	463	86	45	60	76		16	20,600	1.31	6,180	2.56	8	8
	4:20	414	46	436	82	46	60	96		21	10,200	0.79	4,170	2.23	9	4

Table 6.4 (continued)

		Flow Rates, ml/min							Analysis							
		Sorption Section			Elution Section				Feed	Sorption Waste	Elution	Waste	Product			
Date	Time	Feed	Wash	Waste	Eluant	Wash	Waste	Product	Gr γ, c/ml.min	Gr γ, c/ml.min	Gr γ, c/ml.min	H^+, M	Gr γ, c/ml.min	H^+, M	Na^+ ppm	Al ppm
9/6/57	9:30 am	Start														
	10:30	395	45	458	86	45	55	83		28	26,900	0.62	3,020	2.16	186	4
	11:30	405	43	463	83	45	55	86		40	17,300	0.92	7,020	2.40	322	5
	12:30 pm	378	80	483	71	76	61	80		45	14,100	0.92	1,380	1.40	1,520	3

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