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Development of Low-Level Liquid-Waste-Treatment Systems: April-September 1981

Melvin K. Williams, C. Mark Colvin and William H. Bond

March 5, 1982

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MOUND FACILITY
Miamisburg, Ohio 45342

operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company

for the
U. S. DEPARTMENT OF ENERGY
Contract No. DE-AC04-76-DP00053

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Foreword

Under the sponsorship of the DOE Division of Waste Management, Production and Reprocessing, and the direction of the Idaho Operations Office which is responsible for the management of Low Level Waste Programs, Mound is responsible for the development and demonstration of separation methods for removing radionuclides from intermediate-level and low-level liquid processing wastes.

This report is submitted by W. T. Cave, Director, Nuclear Operations, and B. R. Kokenge, Manager, Nuclear Technology, from contributions prepared by members of the Nuclear Waste Technology Section, R. R. Jaeger, Manager, and the Liquid Volume Reduction Technology Group, W. H. Bond, Leader.

To provide an easier understanding of the relationship of the work described herein to the entire project, a work breakdown structure and FY-1981 milestone chart are provided.

Previous reports on this project are listed below:

October 1978-March 1979	MLM-2611
April-September 1979	MLM-2684
October 1979-March 1980	MLM-2735
April-Steptember 1980	MLM-2795
October 1980-March 1981	MLM-2869

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Abstract

The pilot plant reverse osmosis system was demonstrated to be effective in removing large percentages of cobalt-60, iodine-125, and a mixture of cesium-137, cobalt-60, and iodine-125 from two types of aqueous streams. The effectiveness of three membrane porosities, 0, 50, and 97% salt rejection, were explored with each isotope. The 97% salt rejection membrane was the most effective in each experiment. Removals as high as 97.5% of the cobalt, 92.9% of the iodine, and 95.1% of the combined isotopes were achieved.

The effect of possibly interfering factors on the adsorbence of cobalt-60 and iodine-129 on selected ion exchange resins was investigated. The factors thought to affect cobalt-60 adsorption were $[OH^-]$, $[NH_4^+]$, and $[SO_3^=]$. None of the seven factors investigated had any effect on iodine-129 adsorption.

Cesium-137 was removed from a 4,600-gal aqueous waste containing a large amount of sodium hydroxide by treatment with sodium tetraphenyl boron. The cesium concentration of the supernatant portion was reduced from 570 to 4 counts/min/ml.

Introduction

This report is organized to conform to the Work Breakdown Structure (WBS) for the Ultrafiltration and Adsorbents program. A copy of the WBS is shown in Figure 1. Figure 2 is the FY-1981 Milestone Chart for the Program.

3.1.1.1 Reverse Osmosis Pilot Plant Demonstration

C. Mark Colvin

The term "reverse osmosis" is derived from osmosis, the natural phenomenon which occurs in living cell membrances. Normal osmosis occurs when water passes from a less concentrated solution to a more concentrated solution through a semipermeable membrance. A certain amount of potential energy exists between the solutions on either side of the semipermeable membrane. Water will flow because of this energy difference from the less concentrated to the more concentrated solution until the system is in equilibrium. The application of pressure to the concentrated solution will stop the transport of water across the membrance when the applied pressure equals the apparent osmotic pressure between the two solutions. The apparent osmotic pressure is a measurement of the potential energy difference between the two solutions. As more pressure is applied to the more concentrated solution,

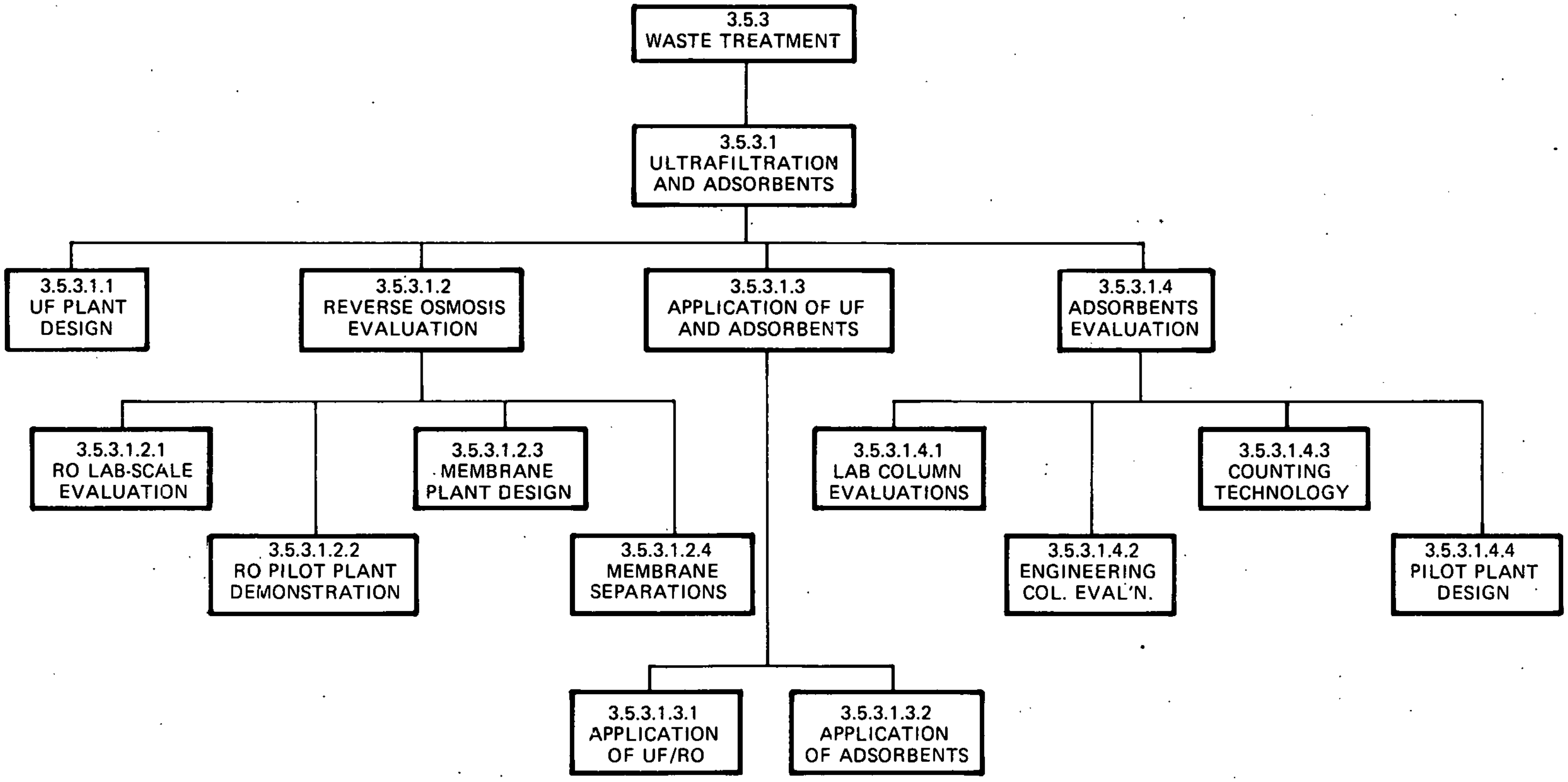

FIGURE 1 - Work breakdown structure.

the water will begin to flow from the concentrated solution to the less concentrated solution. The rate of water transport is a function of the pressure applied and the area of the membrane under pressure. The absolute osmotic pressure is the potential energy difference between any solution and pure water.

Equipment

The reverse osmosis pilot plant, which was fabricated by Osmonics, Inc., of Minneapolis, Minnesota, consists of a transfer pump; two feed pumps connected in series and capable of a combined pressure up to 600 psig; all necessary monitoring equipment such as flow meters, pressure gauges, pH meter, and thermometer; and three types of membrances than can be used either in series or separately. Figure 3 is a flow diagram of the reverse osmosis (RO) unit. The three membranes are: Osmonics Pu-192-43-SS-O-(PS)-8WPT polysulfone membrane rated at 0% sodium chloride rejection; Osmonics Pu-192-43-SS-50-8WPT cellulose acetate (CA) membrane rated at 50% sodium chloride rejection; and Osmonics Pu-192-43-SS-97-8WPT cellulose acetate membrane rated at 97% sodium chloride rejection. Hereafter, in this report, these membranes will be referred to as 0, 50, and 97 membranes, respectively.

The 0 membrane, it should be pointed out, is an ultrafiltration membrane, not a reverse osmosis membrane. A reverse osmosis membrane is capable of rejection or selective retention of ionic impurities. The ionic impurities are repelled by the membrane and restricted from passing through the membrane pore. The size of the ionic impurity is normally smaller than the membrane pore. RO has become generally accepted as the removal of ionic impurities from water by means of a membrane. The rejection or selective retention of nonionic impurities such as organics and emulsified materials has become known as ultrafiltration (UF). Membranes that remove impurities based on size are classified as ultrafiltration membranes. In general, a UF membrane has become commonly accepted as a membrane that does not reject ionic materials. The 0 membrane, because ionic rejection was attempted at operating pressures associated with reverse osmosis, will be referred to as a RO membrane. The fact that the 0 membrane is fabricated of polysulfone, which is tolerant to solutions ranging in pH from 0.5 to 12.5 and is resistant to temperatures in excess of 180°F, makes this a potentially useful membrane. The maximum suggested operating pressure is 200 psig. The manufacturer's suggested operating pressure is 100 psig, although polysulfone membranes tend to compact about 50 psig. The 0 membrane has an average pore size of 15 Å and a molecular weight cutoff of 1000 for organics. The 0 membrane is not effective in salt separation that is dependent on ionization.

Both the 50 and 97 membranes are constructed of cellulose acetate and should be used only in the pH range from 3 to 6. The cellulose acetate membranes vary in maximum operating pressure yet, unlike the polysulfone membrane, the cellulose acetate membranes resist compaction at high pressures. The maximum suggested long-term operating pressure is 300 psig, and the recommended operating pressure is 200 psig for the 50 membrane. The 50 membrane has a molecular weight cutoff

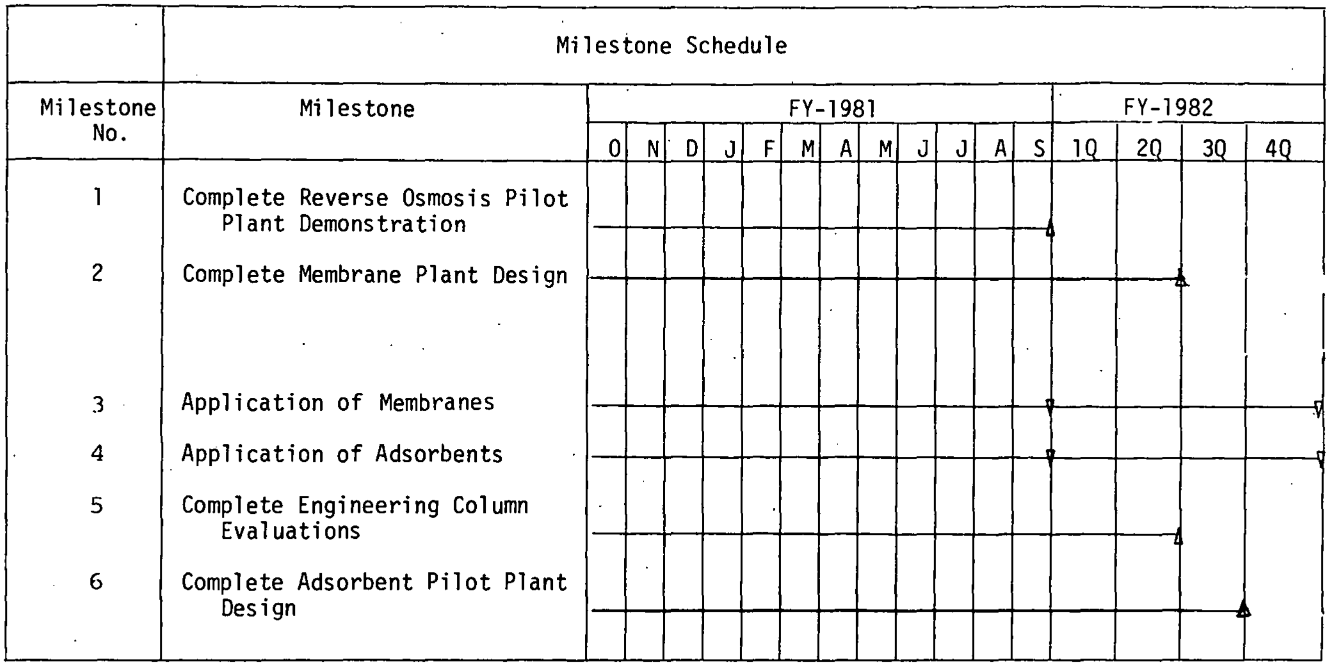

FIGURE 2 - FY-1981 Milestone Chart.

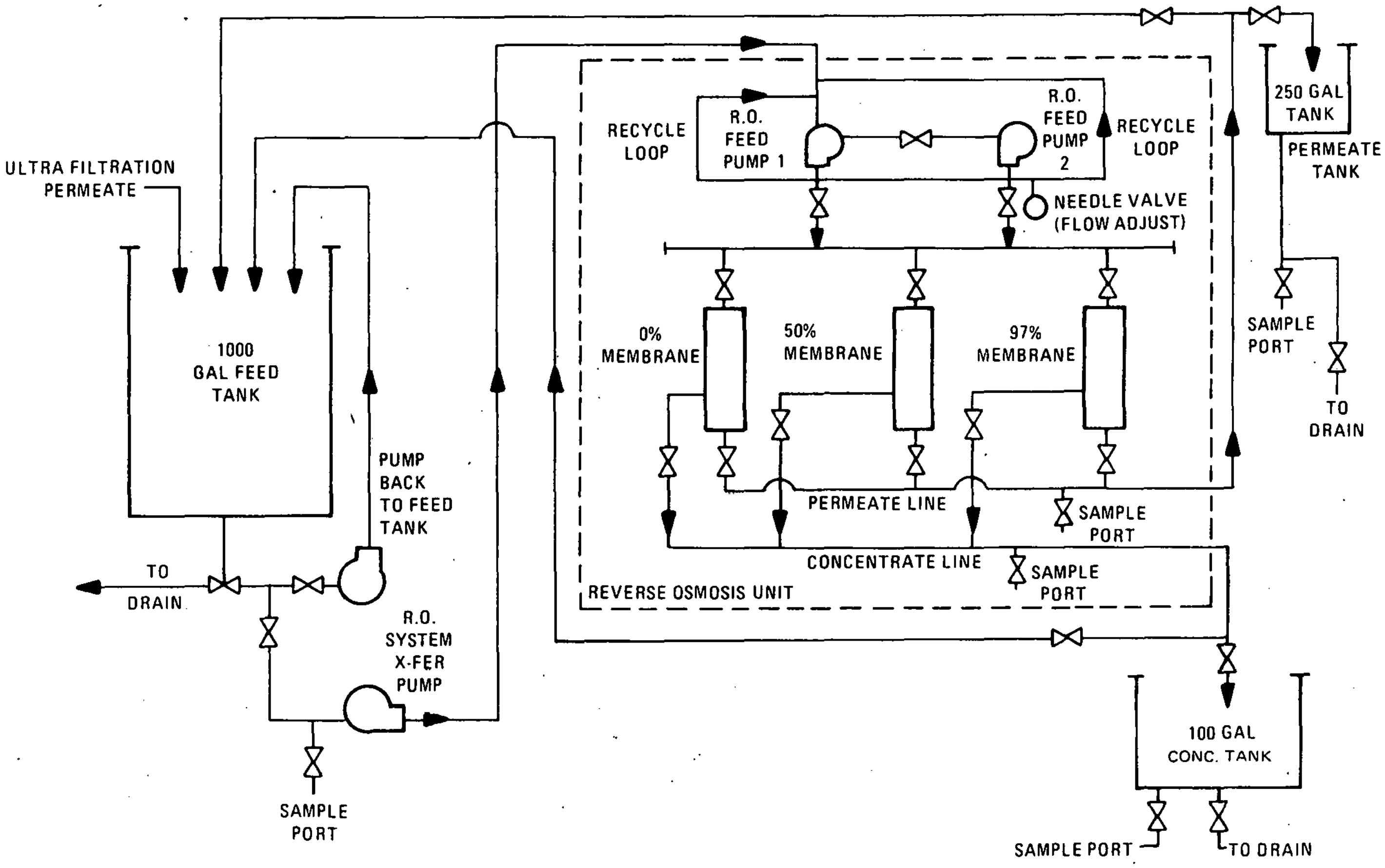

FIGURE 3 - Flow diagram - pilot plant reverse osmosis system.

of 600 for organics, and an average pore size of 11 Å. For the 97 membrane, the maximum suggested operating pressure is 800 psig, and the suggested operating pressure for low compaction is 400 psig. One of the applications of the 97 membrane is the desalination of sea water in which high pressures are necessary to overcome the osmotic pressure of the brine solution. The 97 membrane has a molecular weight cutoff of 200 for organics and an average pore size of 5 Å. The removal of radioactive elements is listed as one of the typical uses of the 97 membrane [1].

Experimental procedure

A series of tests was performed with the reverse osmosis (RO) pilot plant using the isotopes cobalt-60 and iodine-125 to determine the capability of the three types of reverse osmosis membranes to reject the isotopes in varied concentrations and in a simulated waste stream. Tests were also performed with the isotopes cesium-137, cobalt-60, and iodine-125 mixed in equal concentrations in tap water and a simulated waste solution. The procedure followed in these experiments is listed below.

Step 1 The RO feed tank was filled to ∼100 gal with tap water and a known amount of the radionuclide(s) was added.

Step 2 The pH was adjusted to and maintained at between 4.5 and 5.5 with additions of concentrated nitric acid.

Step 3 The feed solution was agitated for at least 3 hr to allow the isotope(s) to come to equilibrium.

Step 4 A 100-ml feed sample was taken, and the pH was measured with a calibrated pH meter.

Step 5 The RO unit was set to recycle: the two streams formed by the membrane are returned to the feed tank where they are mixed with the remaining feed solution; then the solution was pumped through the 0 membrane.

Step 6 The recommended operating pressure was obtained and maintained. (The recommended operating pressures are stated earlier in this report.)

Step 7 The feed solution was allowed to recycle for ∼45 min, then a 100-ml sample of the permeate was taken, and two additional 100-ml permeate samples were taken at 15-min intervals.

Step 8 Steps 4 through 7 were repeated with the 50 and 97 membranes in respective order.

The membranes were tested with varied isotope concentrations to determine if rejection is a function of concentration. In this report, rejection will be presented as a percentage, the formaula for which is,

$$\% \text{ R} = \frac{\text{Feed conc.} - \text{permeate conc.}}{\text{feed conc.}} \times 100 .$$

The membranes were tested at low, medium, and high isotope concentrations. The membranes were also tested with a simulated waste stream to observe the effects of ionic impurities on the isotope(s) rejection.

The simulated waste stream included only impurities that might be dissolved in a worst case actual waste stream after pretreatment by ultrafiltration and anion exchange. An analysis of any waste stream to be treated with RO is necessary to determine what type of pretreatment, if any, is needed. A possible general process for treating a waste stream that has a great deal of impurities would include the following steps:

Step 1 The solution would be adjusted to a basic pH and run through an ultrafiltration system to remove the undissolved solids.

Step 2 The solution would be neutralized and run through an anion exchange resin to remove the sulfates and the carbonates, which could cause scale damage to the membranes. This step would also remove the $PO_4^{\equiv}$ and $HPO_4^{=}$.

Step 3 The solution would be adjusted to ∿pH 5 and passed through a reverse osmosis system.

Step 4 Cation exchange could be used as a polishing step.

The impurities used in the simulated waste stream were the ions, from a worst case waste stream, that would remain in solution after Step 2. The ions used in the simulated waste stream are listed in Table 1.

Results

The results of the single isotope experiments are listed in Tables 2 to 5 for cobalt-60, and Tables 6 to 9 for iodine-125. The results of mixed isotopes experiments are listed in Tables 10 and 11. The tables are displayed for evaluation of the performance, by comparison, of the 0, 50, and 97 membranes with the title isotope(s). The numbers under the heading of $\bar{x}$ feed and $\bar{x}$ permeate are the average counts of the samples taken during the runs. Presented in the lower section of the tables are the average percent rejection, the standard deviation, and the confidence limits for each membrane. Uncertainties are reported at the 90% confidence level. All samples taken throughout the duration of the experiment were counted on a Packard 460 CD liquid scintillation spectrometer. The results listed in the tables for the mixed isotopes experiments are the percentage of the total beta activity rejected by the membrane.

There are two fundamental mechanisms of rejection at work in the reverse osmosis process. Salt rejection is one of these mechanisms. Salt rejection uses the electrical charge carried by ions. The magnitude of this charge is a function of the valence of the ion. Whether cationic or anionic, the ion is, in general, repelled from the surface of the membrane a distance proportional to its valence ($Al^{+3} > Ca^{+2} > Na^{+}$). The reason the ions are repulsed from the surface of the membrane is considered to be an electrostatic "dipole" effect that is set up between the charged ion and the surface of the membrane by virtue of a "mirror" effect [2]. In other words, the charge on the ion sets up, or induces, an equal

Table 1 - CONCENTRATION OF IONS IN SIMULATED WASTE STREAM

Cations			Anions		
Conc. (g/L)	Cation	Compound Used	Conc. (g/L)	Anion	Compound Used
1.5	Ca^{++}	$CaCl_2$	8.7	Cl-	(All Cations)
3.0	Na^{+}	NaCl (NaI)			
0.01	Ba^{++}	$BaCl_2$	0.005	I^-	NaI
0.4	Mg^{++}	$MgCl_2$	0.005	Br^-	KBr
0.3	K^{+}	KCl (KF)		NO_3 via HNO_3	Acidifier

Na_2HPO_4 - 0.1 g/L

Table 2 - RESULTS OF RO MEMBRANE TEST WITH COBALT-60 LOW CONCENTRATION

Run No.	$\bar{X}$ Feed β (counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	324	263	18.8	35	89.4	12	96.3
2	204	325	0	35	88.1	15	94.8
3	316	322	0	31	90.2	8	97.4
4	314	342	0	37	88.2	12	96.2
$\bar{X}$ Percent Rejection			4.7		89.0		97.0
Standard Deviation			9.4		1.0		0.7
Confidence Limits at 90% Probability Level			4.7 ± 11.1		89.0 ± 1.2		97.0 ± 0.8

Table 3 - RESULTS OF RO MEMBRANE TEST WITH COBALT-60 MEDIUM CONCENTRATION

Run No.	$\bar{X}$ Feed β (Counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	6488	6259	3.6	413	93.6	227	96.5
2	6453	6576	0	334	94.8	149	97.7
3	6439	7546	0	293	95.5	116	98.2
4	6448	6233	3.5	280	95.7	118	98.2
$\bar{X}$ Percent Rejection			1.8		94.9		97.7
Standard Deviatiom			2.1		1.0		0.8
Confidence Limits at 90% Probability Level			1.8 ± 2.5		94.9 ± 1.2		97.7 ± 0.9

Table 4 - RESULTS OF RO MEMBRANE TEST WITH COBALT-60 HIGH CONCENTRATION

Run No.	$\bar{X}$ Feed β (counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	12612	11288	10.5	572	95.5	462	96.3
2	11297	10799	4.4	519	95.4	247	97.8
3	11473	11351	1.2	544	95.3	246	97.9
4	11672	11291	3.3	547	95.3	242	97.9
$\bar{X}$ Percent Rejection			4.9		95.4		97.5
Standard Deviation			4.0		0.1		0.8
Confidence Limits at 90% Probability Level			4.9 ± 4.7		95.4 ± 0.1		97.5 ± 0.9

Table 5 - RESULTS OF RO MEMBRANE TEST WITH COBALT-60 SIMULATED WASTE STREAM

Run No.	$\bar{X}$ Feed β (counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	8026	8495	0	1314	83.6	333	95.9
2	7821	8271	0	1264	83.8	387	95.1
3	8186	8192	0	1218	85.1	472	94.2
4	8239	7476	9.3	1426	83.0	424	94.9
$\bar{X}$ Percent Rejection			2.3		83.9		95.0
Standard Deviation			4.7		0.9		0.7
Confidence Limits at 90% Probability Level			2.3 ± 5.5		83.9 ± 1.1		95.0 ± 0.3

Table 6 - RESULTS OF RO MEMBRANE TEST WITH I-125 LOW CONCENTRATION

No.	$\bar{X}$ Feed β (counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane X Permeate β (counts/min/ml)	Percent Rejection
1	361	353	2.1	248	31.3	55	84.9
2	364	340	6.7	238	34.5	57	84.3
3	358	343	4.3	221	38.2	53	85.2
4	356	352	1.2	225	36.8	49	86.2
$\bar{X}$ Percent Rejection			3.6		35.2		85.2
Standard Deviation			2.5		3.0		0.8
Confidence Limits at 90% Probability Level			36 ± 2.9		35.2 ± 3.5		85.2 ± 1.0

Table 7 - RESULTS OF RO MEMBRANE TEST WITH IODINE-125 MEDIUM CONCENTRATION

Run No.	$\bar{X}$ Feed β (counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	3408	3156	7.4	2087	38.8	544	84.1
2	3364	3161	6.1	2262	32.8	544	83.9
3	3418	3438	0	2351	31.4	550	84.0
4	3679	3354	8.9	2354	36.0	533	85.8
$\bar{X}$ Percent Rejection			5.6		34.8		84.5
Standard Deviation			3.9		3.3		0.9
Confidence Limits at 90% Probability Level			5.6 ± 4.6		34.8 ± 3.9		84.5 ± 1.1

Table 8 - RESULTS OF RO MEMBRANE TEST WITH IODINE-125 HIGH CONCENTRATION

No.	$\bar{X}$ Feed β (counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Percent Rejection	97-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	9993	9767	2.3	4818	48.2	677	93.2
2	10106	9768	3.4	5022	49.7	718	92.9
3	100017	9739	2.8	5166	51.5	725	92.8
4	10090	9769	3.2	5202	51.5	752	92.5
$\bar{X}$ Percent Rejection			2.9		49.8		92.9
Standard Deviation			0.5		1.6		0.3
Confidence Limits at 90% Probability Level			2.9 ± 0.6		49.8 ± 1.9		92.9 ± 0.3

Table 9 - RESULTS OF RO MEMBRANE TEST WITH I-125 SIMULATED WASTE STREAM

Run No.	$\overline{X}$ Feed β (counts/min/ml)	0-Membrane $\overline{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\overline{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\overline{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	9568	9555	0.2	5968	37.7	1573	83.6
2	9557	9524	0.6	5888	38.4	1491	84.4
3	9510	9400	1.2	5861	38.4	1503	84.2
4	9229	9131	1.0	5857	36.5	1498	83.8
$\overline{X}$ Percent Rejection			0.8		37.8		84.0
Standard Deviation			0.4		0.9		0.4
Confidence Limits at 90% Probability Level			0.8 ± 0.4		37.8 ± 1.1		84.0 ± 0.4

Table 10 - RESULTS OF RO MEMBRANE TEST WITH MIXED ISOTOPES (Cs-137, Co-60, I-125)

Run No.	$\overline{X}$ Feed β (counts/min/ml)	0-Membrane $\overline{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\overline{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\overline{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	44299	44739	0	21402	51.7	2249	94.9
2	44350	36510	17.7	21565	51.4	2124	95.2
3	44115	30766	30.3	21414	51.5	2148	95.1
4	43691	36569	16.5	21368	51.1	2178	95.0
$\overline{X}$ Percent Rejection			16.1		51.4		95.1
Standard Deviation			12.4		0.3		0.1
Confidence Limits at 90% Probability Level			16.1 ± 14.6		51.4 ± 0.3		95.1 ± 0.2

Table 11 - Results of RO MEMBRANE TEST WITH MIXED ISOTOPES (Cs-137, Co-60, I-125) SIMULATED WASTE STREAM

Run No.	$\bar{X}$ Feed β (counts/min/ml)	0-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	50-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection	97-Membrane $\bar{X}$ Permeate β (counts/min/ml)	Percent Rejection
1	40589	39086	3.7	23035	43.3	5263	87.1
2	40245	38849	3.5	23089	42.7	4986	87.7
3	40299	39197	2.8	23174	42.9	5257	87.0
4	39903	39167	1.9	23290	41.7	5246	86.9
$\bar{X}$ Percent Rejection			3.0		42.7		87.2
Standard Deviation			0.8		0.7		0.4
Confidence Limits at 90% Probability Level			3.0 ± 1.0		42.7 ± 1.4		87.2 ± 0.4

and like charge on the membrane surface which causes a force or repulsion between the membrane and the charged ion. The second mechanism is that of organic rejection in which dissolved organics are rejected by a screening, or sieving, mechanism. The rejection of any given organic molecule is a function of membrane pore size, size of the molecule, and the geometry of the molecule. Isotopes complexed with organics of large molecular weight will tend to act like the organics they are complexed with.

The 0 membrane did not perform well with the single isotopes in varied concentrations or in a simulated waste stream. The results of cobalt-60 tests with the 0 membrane, expressed as percent rejection, are as follows: the low concentration, 4.7% ± 11.1% (Table 2); the medium concentration, 1.8% ± 2.5% (Table 3); the high concentration, 4.9% ± 4.7% (Table 4); and the simulated waste stream, 2.3% ± 5.5% (Table 5). The results of the cobalt-60 tests with the 50 membrane are as follows: the low concentration, 89.0% ± 1.2% (Table 2); the medium concentration, 94.9% ± 1.2% (Table 3); the high concentration, 95.4% ± 0.1% (Table 4); and the simulated waste stream 83.9% ± 1.1% (Table 5). The results of the cobalt-60 tests with the 97 membrane are as follows: the low concentration, 97.0% ± 0.8% (Table 2); the medium concentration, 97.7% ± 9% (Table 3); the high concentration, 97.5% ± (Table 4); and the simulated waste stream, 95.0% ± 0.8% (Table 5).

The results of the iodine-125 tests with the 0-membrane are as follows: the low concentration, 36% ± 2.9% (Table 6); the medium concentration, 5.6% ± 4.6% (Table 7); the high concentration, 2.9% ± 0.6% Table 8); and the simulated waste stream 0.8% ± 0.4% (Table 9). The results of the iodine-125 tests with the 50 membrane are as follows: the low concentration, 35.2% (Table 6); the medium concentration, 34.8% ± 3.9% (Table 7); the high concentration, 49.8% ± 1.9% (Table 8); and the simulated waste stream, 37.8% ± 1.1% (Table 9). The results of the iodine-125 tests with the 97 membrane are as follows: the low concentration, 85.2% ± 1.0% (Table 6); the medium concentration, 84.5% ± 1.1% (Table 7): the high concentration, 92.9% ± 0.3% (Table 8); and the simulated waste stream, 84.0% ± 0.4% (Table 9). The tests with varied isotope concentration demonstrated no significant effect on the isotope rejection by the membrane, although the membrane appeared to perform slightly better at the high concentration.

In the mixed isotopes experiments, the 0 membrane again performed poorly as was anticipated from the results of the single isotopes experiments and the fact that the membrane is rated at 0% rejection of NaCl. The results of the mixed isotopes experiments with the 0 membrane were 16.1% ± 14.6% (Table 10) in tap water and 3.0% ± 1.0% (Table 11) in the simulated waste solution. The results of the mixed isotopes experiments with the 50 membrane were 51.4% ± 0.5% (Table 11) in tap water and 42.7% ± 1.4% (Table 11) in the simulated waste solution. The results of the mixed isotopes experiments with the 97 membrane were 95.1% ± 0.2% (Table 10) in tap water and 87.2% ± 0.4% in the simulated waste solution. In general the membranes performed as anticipated, with the membranes performing somewhat better with the isotope(s) in tap water than in the simulated waste solution.

The results of these experiments indicate that the 97 membrane could be used effectively in volume reduction of cesium-137, cobalt-60, and iodine-125 low-level aqueous wastes.

3.1.4.2 Engineering Column Evaluations

Melvin K. Williams

Cobalt main effects design experiment

The cobalt-60 main effects design as outlined in the previous report [3] was changed by the additon of [Mg++] as an eighth variable. In addition, all upper levels of the variables were changed. The variables and their upper and lower levels are given in Table 12. Some of the upper limits of the variables have been increased and some decreased in reference to the design outlined in the previous report [3].

The worst case of total dissolved solids (TDS) in terms of cations in this experiment was 3587 ppm. This is well above the level of TDS that could be considered economically feasible for ion exchange when the resins are not regenerated but disposed of after using [4]. In terms of equivalents per liter, this worst case was 0.192 equivalents/liter. Over a 15-liter run, this would amount to 2.89 equivalents. Because only about 12 liters of feed were usually used on a run, the number of equivalents actually passed through a resin on the worst case run was 2.31 equivalents. This value exceeds the capacity of most resins; however, 0.981 equivalents of the worst case solution were in the form of Na^+ which should not have affected the sodium form resins. Actual equivalents affecting the resin on the worst case run were 1.33 equivalents. This value did not exceed the capacity of any of the resins used in cobalt-60 main effects design.

The resins used in the cobalt-60 main effects design were: IRC-718, MSC-1, HCR-2W-H, and AG50WX8; all in sodium form.

The constants were: flow rate = 400 ml/min, or ≈0.4 bed volumes per minute, and $[Co^{60}]$ = 10,000 counts/min/ml. The cobalt-60 concentration of 10,000 counts/min/ml is approximately equal to 6.6×10^{-11} moles/liter or 3.99×10^{-6} ppm.

Table 12 - COBALT-60 MAIN EFFECTS DESIGN: VARIABLES AND LIMITS

VARIABLE	X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8
Variable Name	$[Ni^{++}]$	$[Ca^{++}]$	$[Mg^{++}]$	$[OH^-]$	$[NH_4^+]$	$[CO_3^{=}]$	$[PO^{\equiv}]$	$[SO_3^{=}]$
Upper Level (+)	1.0 ppm	1500 ppm	400 ppm	ph-10	50 ppm	1500 ppm	1000 ppm	10 ppm
Lower Level (-)	R.O. Water	R.O. Water	R.O. Water	ph-3	R.O. Water	R.O. Water	R.O. Water	R.O Water

PROCEDURE

The first step of the procedure used in this experiment was the preparation of the feed solutions for a particular run. The feed solution composition was determined by the experimental design (Figure 4). At these variable concentrations, some feed solutions were expected to produce precipitates, and in fact, four of them (runs 3,5,6,9) did. In those cases when precipatation did occur, filtering removed the precipitate before the solution was spiked with cobalt-60. This filtering step was necessary because some precipitates, such as $Ca_3(PO_4)_2$, are good adsorbents for cobalt, and if the cobalt were present during a precipitation, it could be coprecipitated. Also a precipitate in the column feed solution would interfere with the ion exchange resin and possibly clog the colum frits. After the feed solutions were prepared, they were sampled and then pumped through the sodium form resin which had been washed, regenerated, backwashed, and measured to a column volume of one liter. Flow rate was adjusted on the wash or pregeneration steps to ≈ 400 ml/min. Flow rate was not a critical factor [5] but was still calibrated as closely as possible. Four samples were taken, spread evenly over the duration of each run. The samples were then prepared for scintillation counting in a Packard 460CD liquid scintillation counter.

T Trial	Run Order	X_1 $[Ni^{++}]$	X_2 $[Ca^{++}]$	X_3 $[Mg^{++}]$	X_4 $[OH^-]$	X_5 $[NH_4^+]$	X_6 $[Co_3^=]$	X_7 $[PO_4^=]$	X_8 $[SO_3^=]$	X_9	X_{10}	X_{11}	Y % Co^{60} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	
2	11	-	+	-	-	-	+	+	+	-	+	-	
3	8	+	-	-	-	+	+	+	-	+	-	-	
4	10	-	-	-	+	+	+	-	+	-	-	+	
5	5	-	-	+	+	+	-	+	-	-	+	-	
6	6	-	+	+	+	-	+	-	-	+	-	-	
7	1	+	+	+	-	+	-	-	+	-	-	-	
8	3	+	+	-	+	-	-	+	-	-	-	+	
9	12	+	-	+	-	-	+	-	-	-	+	+	
10	2	-	-	-	-	+	-	-	-	+	+	+	
11	7	+	-	-	+	-	-	-	+	+	+	-	
12	9	+	+	+	+	+	+	+	+	+	+	+	

$\Sigma XY =$

$\frac{\Sigma XY}{T/2} =$

$\frac{\Sigma XY}{T/2} /2 = b =$

FIGURE 4 - Cobalt-60 main effects design.

RESULTS

During the analysis of the data, several data transforms were tried to determine which would be the best to use. Data transforms are used in experimental designs to compress the data in situations where the range is very large or where upper or lower limits in the natural data cause a bunching or clustering around endpoint values. In this experiment, if effluent counts were used, the range is very large (2.3 to 9206.6). Because there were only 12 actual data points, and their values were added and subtracted during the analysis of the design, it was important that they be of similar magnitude and weighted closely. Otherwise, one large value could totally dominate the calculation and thus produce misleading results. In the past, percent of the isotope removed from the solution has been the transform used. The percent transform compresses the range to 0 to 100. Other transforms tried, were: ln effluent counts, ln % removed, $\ln\left(\frac{\text{feed counts}}{\text{effluent counts}}\right)$ and $\sin^{-1}\left(\sqrt{\frac{\%}{100}}\right)$. The transform $\sin^{-1}\left(\sqrt{\frac{\%}{100}}\right)$ was suggested by the experimental design group as the transform of choice for this type of experiment. The arc sin transform produced a scale slightly compressed over the percent transform. The natural log transforms were all very compressed (See Table 13).

The results of the data analyses varied somewhat, depending on the data transform used. All analyses indicated a very large interaction of two or more variables. Variable interactions are not measured by main effects designs and tend to have their effect spread throughout all variables. Because of this large interaction, the less discriminating (less compreseed) data transforms show no effects, whereas the more discriminating (more compressed) transforms, in some cases, do show effects. The most useful of the data transforms was the natural log of the effluent counts. Therefore, both the data analyzed using a percent of cobalt-60 removed and ln of the effluent counts will be discussed.

Table 13 - DATA TRANSFORMS AND NATURAL DATA COMPARISON

TRANSFORM		RANGE ON DATA FROM AG50WX8		
		Run 3		Run 4
Natural Data:	Effluent counts/min/ml	3.2	to	8836.1
	In (Effluent counts/min/ml)	1.16	to	9.09
	% Co^{60} Removed	99.97	to	1.64
	ln (% Co^{60} Removed	4.60	to	0.49
	$\ln\left[\frac{\text{(Feed counts/min/ml)}}{\text{(Effluent counts/min/ml)}}\right]$ = ln(DF)	8.07	to	0.02
	$\sin^{-1}\left(\sqrt{\frac{\%}{100}}\right)$	89.01	to	7.36

Figures 5, 6, 7, and 8 are the experimental design analyses using percent of cobalt-60 removed as the data transform. Figure 5 is the data analysis for AG50WX8. Runs 9 and 10 show some large interaction, but the b's (bottom row) with confidence limits of ± 27.18 all contain zero. Therefore, at the 95% confidence level, there are no single factor effects. Figure 6 is the data analysis for MSC-1. Again, runs 9 and 10 show a large interaction effect, but all the b's with confidence limits of ± 20.45 contain zero. Figure 7 is the data analysis for IRC-718. The very large interaction is apparent in run 10. The confidence limits are ± 23.22. Because all the b's with the ± 23.22 confidence limits contain zero, there are no single factor effects. IRC-718 gave very different results from the other cation exchange resins and, in general, was not as successful in removing cobalt from aqueous solution. Figure 8 is the data analysis for HCR-2S-H. Again, run 10 shows the large variable interaction, and the b confidence limits are large enough to place zero in the interval for all the b's. Run 9 does not show the large interaction which is seen in run 10. Since run 9 was filtered, it is believed that one of the factors needed for the large interaction effect must have been removed.

The data analyses using the transform ln (effluent counts) are presented in Figures 9 through 12. Figure 9 is the analysis of AG50WX8. The large interaction is apparent in runs 9 and 10. The confidence limits for the b's are ± 1.29. These confidence limits show $[SO_3^=]$ as a factor and also give close values for $[OH^-]$, $[NH_4^+]$ and $[SO_3^=]$. Notice in comparison to Figure 5 where $[OH^-]$, $[NH_4^+]$, $[CO_3^=]$ and $[SO_3^=]$ all have close b values, with the more compressed transform, $[CO_3^=]$ is no longer in the same range with the other factors.

T Trial	Run Order	X_1 $[Ni^{++}]$	X_2 $[Ca^{++}]$	X_3 $[Mg^{++}]$	X_4 $[OH^-]$	X_5 $[NH_4^+]$	X_6 $[Co_3^=]$	X_7 $[PO_4^{\equiv}]$	X_8 $[SO_3^=]$	X_9	X_{10}	X_{11}	Y % Co^{60} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	99.89
2	11	-	+	-	-	-	+	+	+	-	+	-	99.80
3	8	+	-	-	-	+	+	+	-	+	-	-	99.97
4	10	-	-	-	+	+	+	-	+	-	-	+	1.64
5	5	-	-	+	+	+	-	+	-	-	+	-	99.32
6	6	-	+	+	+	-	+	-	-	+	-	-	99.78
7	1	+	+	+	-	+	-	-	+	-	-	-	98.00
8	3	+	+	-	+	-	-	+	-	-	-	+	99.85
9	12	+	-	+	-	-	+	-	-	-	+	+	99.79
10	2	-	+	-	-	+	-	-	-	+	+	+	99.89
11	7	+	-	-	+	-	-	-	+	+	+	-	99.68
12	9	+	+	+	+	+	+	+	+	+	+	+	20.18
$\Sigma XY =$		17.15	17.22	16.13	176.89	-179.79	-175.47	20.23	-179.41	20.99	19.53	-175.31	
$\frac{\Sigma XY}{T/2} =$		2.86	2.87	2.64	-29.48	- 29.97	- 29.25	3.37	- 29.9	3.50	3.26	- 29.22	
$\frac{\Sigma XY}{T/2}/2 =$		1.43	1.43	1.34	-14.74	- 14.98	- 14.62	1.69	- 14.95	1.75	1.63	- 14.61	

s = 29.61
t = 3.18
b ± 27.18

FIGURE 5 - Cobalt-60 main effects design (AG50WX8).

T Trial	Run Order	X_1 [Ni^{++}]	X_2 [Ca^{++}]	X_3 [Mg^{++}]	X_4 [OH^-]	X_5 [NH_4^+]	X_6 [$CO_3^=$]	X_7 [$PO_4^\equiv$]	X_8 [$SO_3^=$]	X_9	X_{10}	X_{11}	Y % Co^{60} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	99.69
2	11	-	+	-	-	-	+	+	+	-	+	-	99.70
3	8	+	-	-	-	+	+	+	-	+	-	-	99.92
4	10	-	-	-	+	+	+	-	+	-	-	+	28.98
5	5	-	-	+	+	+	-	+	-	-	+	-	99.22
6	6	-	+	+	+	-	+	-	-	+	-	-	99.10
7	1	+	+	+	-	+	-	-	+	-	-	-	90.73
8	3	+	+	-	+	-	-	+	-	-	-	+	99.88
9	12	+	-	+	-	-	+	-	-	-	+	+	99.86
10	2	-	+	-	-	+	-	-	-	+	+	+	99.91
11	7	+	-	-	+	-	-	-	+	+	+	-	99.18
12	9	+	+	+	+	+	+	+	+	+	+	+	26.14
ΣXY =		-10.89	-11.39	-12.83	-137.31	-152.51	-134.91	6.79	-153.47	5.57	6.71	-133.39	
$\frac{\Sigma XY}{T/2}$ =		- 1.82	- 1.90	- 2.14	- 22.89	- 25.42	- 22.49	1.13	- 25.58	0.93	0.95	- 22.23	
$\frac{\Sigma XY}{T/2}$ /2 =		- 0.91	- 0.95	- 1.07	- 11.44	- 12.71	- 11.24	0.57	- 12.79	0.46	0.48	- 11.12	

s = 22.28
t = 3.18
b ± 20.45

FIGURE 6 - Cobalt main effects design (MSC-1).

T Trial	Run Order	X_1 [Ni^{++}]	X_2 [Ca^{++}]	X_3 [Mg^{++}]	X_4 [OH^-]	X_5 [NH_4^+]	X_6 [$Co_3^=$]	X_7 [$PO_4^\equiv$]	[$SO_3^=$]	X_9	X_{10}	X_{11}	Y % Co^{60} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	99.83
2	11	-	+	-	-	-	+	+	+	-	+	-	99.98
3	8	+	-	-	-	+	+	+	-	+	-	-	94.30
4	10	-	-	-	+	+	+	-	+	-	-	+	11.19
5	5	-	-	+	+	+	-	+	-	-	+	-	84.94
6	6	-	+	+	+	-	+	-	-	+	-	-	74.50
7	1	+	+	+	-	+	-	-	+	-	-	-	97.37
8	3	+	+	-	+	-	-	+	-	-	-	+	76.30
9	12	+	-	+	-	-	+	-	-	-	+	+	99.83
10	2	-	+	-	-	+	-	-	-	+	+	+	55.35
11	7	+	-	-	+	-	-	-	+	+	+	-	99.60
12	9	+	+	+	+	+	+	+	+	+	+	+	68.42
ΣXY =		110.03	-17.77	88.17	-131.71	-138.47	-65.17	85.93	-8.32	22.39	54.63	-139.77	
$\frac{\Sigma XY}{T/2}$ =		18.34	- 2.96	14.70	- 21.95	- 23.08	-10.86	14.32	-1.47	3.73	9.11	- 23.3	
$\frac{\Sigma XY}{T/2}$ /2 =		9.17	- 1.48	7.35	- 10.98	- 11.54	- 5.43	7.16	-0.74	1.87	4.55	- 11.65	

s = 25.29
t = 3.18
b ± 23.22

FIGURE 7 - Cobalt-60 main effects design (IRC-718).

T Trial	Run Order	X_1 $[Ni^{++}]$	X_2 $[Ca^{++}]$	X_3 $[Mg^{++}]$	X_4 $[OH^-]$	X_5 $[NH_4^+]$	X_6 $[Co_3^=]$	X_7 $[PO_4^\equiv]$	X_8 $[SO_3^=]$	X_9	X_{10}	X_{11}	Y % Co^{60} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	99.91
2	11	-	+	-	-	-	+	+	+	-	+	-	99.86
3	8	+	-	-	-	+	+	+	-	+	-	-	99.96
4	10	-	-	-	+	+	+	-	+	-	-	+	1.41
5	5	-	-	+	+	+	-	+	-	-	+	-	99.49
6	6	-	+	+	+	-	+	-	-	+	-	-	98.65
7	1	+	+	+	-	+	-	-	+	-	-	-	98.27
8	3	+	+	-	+	-	-	+	-	-	-	+	99.46
9	12	+	-	+	-	-	+	-	-	-	+	+	99.83
10	2	-	+	-	-	+	-	-	-	+	+	+	99.92
11	7	+	-	-	+	-	-	-	+	+	+	-	99.98
12	9	+	+	+	+	+	+	+	+	+	+	+	98.78
ΣXY =		97.02	94.38	94.36	-100.00	-99.84	-95.82	99.42	-99.12	98.86	100.18	-96.88	
$\frac{\Sigma XY}{T/2}$ =		16.17	15.73	15.73	- 16.67	-16.64	-16.42	16.57	-16.57	16.48	16.70	-16.15	
$\frac{\Sigma XY}{T/2}$ /2 =		8.09	7.87	7.86	-8.33	- 8.32	- 8.21	8.29	- 8.26	8.24	8.35	- 8.07	

s = 28.48

t = 3.18

b ± 26.14

FIGURE 8 - Cobalt-60 main effects design (HCR-2W-H).

T Trial	Run Order	X_1 $[Ni^{++}]$	X_2 $[Ca^{++}]$	X_3 $[Mg^{++}]$	X_4 $[OH^-]$	X_5 $[NH_4^+]$	X_6 $[Co_3^=]$	X_7 $[PO_4^\equiv]$	X_8 $[SO_3^=]$	X_9	X_{10}	X_{11}	Y c/m/ml Eff	ln Counts
1	4	-	-	+	-	-	-	+	+	+	-	+	11.8	2.47
2	11	-	+	-	-	-	+	+	+	-	+	-	18.6	2.92
3	8	+		-	-	+	+	+	-	+	-	-	3.2	1.16
4	10	-	-	-	+	+	+	-	+	-	-	+	8836.1	9.09
5	5	-	-	+	+	+	-	+	-	-	+	-	68.2	4.22
6	6	-	+	+	+	-	+	-	-	+	-	-	21.7	3.08
7	1	+	+	+	-	+	-	-	+	-	-	-	186.9	5.23
8	3	+	+	-	+	-	-	+	-	-	-	+	12.1	2.49
9	12	+	-	+	-	-	+	-	-	-	+	+	19.9	2.99
10	2	-	+	-	-	+	-	-	-	+	+	+	12.1	2.49
11	7	+	-	-	+	-	-	-	+	+	+	-	33.0	3.50
12	9	+	+	+	+	+	+	+	+	+	+	+	7939.6	8.98
ΣXY =		0.08	1.76	5.32	14.10	13.72	7.82	-4.14	15.76	-5.26	1.58	6.36		
$\frac{\Sigma XY}{T/2}$ =		0.01	0.29	0.89	2.35	2.29	1.30	-0.69	2.63	-0.88	0.26	1.06		
$\frac{\Sigma XY}{T/2}$ /2 =		0.01	0.15	0.44	1.18	1.14	0.65	-0.35	1.31	-0.44	0.13	0.53		

s = 1.40

t = 3.18

b ± 1.29

FIGURE 9 - Cobalt-60 main effects design (AG50WX8).

T Trial	Run Order	X_1 $[Ni^{++}]$	X_2 $[Ca^{++}]$	X_3 $[Mg^{++}]$	X_4 $[OH^{-}]$	X_5 $[NH_4^{+}]$	X_6 $[Co_3^{=}]$	X_7 $[PO_4^{\equiv}]$	X_8 $[SO_3^{=}]$	X_9	X_{10}	X_{11}	Y c/m/ml Eff	ln Counts
1	4	-	-	+	-	-	-	+	+	+	-	+	32.6	3.48
2	11	-	+	-	-	-	+	+	+	-	+	-	29.5	3.38
3	8	+	-	-	-	+	+	+	-	+	-	-	7.6	2.03
4	10	-	-	-	+	+	+	-	+	-	-	+	7042.8	8.86
5	5	-	-	+	+	+	-	+	-	-	+	-	77.1	4.35
6	6	-	+	+	+	-	+	-	-	+	-	-	91.4	4.52
7	1	+	+	+	-	+	-	-	+	-	-	-	885.7	6.79
8	3	+	+	-	+	-	-	+	-	-	-	+	11.6	2.45
9	12	+	-	+	-	-	+	-	-	-	+	+	13.1	2.57
10	2	-	+	-	-	+	-	-	-	+	+	+	9.3	2.23
11	7	+	-	-	+	-	-	-	+	+	+	-	76.23	4.33
12	9	+	+	+	+	+	+	+	+	+	+	+	6860.3	8.83
$\Sigma XY =$		0.18	2.58	7.26	12.86	12.36	6.56	-4.78	17.52	-2.98	-2.44	3.02		s = .81
$\frac{\Sigma XY}{T/2} =$		0.03	0.43	1.21	2.14	2.06	1.09	-0.80	2.92	-0.50	-0.41	0.50		t = 3.18
$\frac{\Sigma XY}{T/2}/2 =$		0.02	0.22	0.61	1.07	1.03	0.55	-0.40	1.46	-0.25	-0.20	0.25		b ± 0.74

FIGURE 10 - Cobalt 60 main effects design (MSC-1).

T Trial	Run Order	X_1 $[Ni^{++}]$	X_2 $[Ca^{++}]$	X_3 $[Mg^{++}]$	X_4 $[OH^{-}]$	X_5 $[NH_4^{+}]$	X_6 $[Co_3^{=}]$	X_7 $[PO_4^{\equiv}]$	X_8 $[SO_3^{=}]$	X_9	X_{10}	X_{11}	Y c/m/ml Eff	ln Counts
1	4	-	-	+	-	-	-	+	+	+	-	+	17.8	2.88
2	11	-	+	-	-	-	+	+	+	-	+	-	2.3	0.83
3	8	+	-	-	-	+	+	+	-	+	-	-	505.9	6.37
4	10	-	-	-	+	+	+	-	+	-	-	+	8531.5	9.05
5	5	-	-	+	+	+	-	+	-	-	+	-	1443.4	7.27
6	6	-	+	+	+	-	+	-	-	+	-	-	2323-1	7.75
7	1	+	+	+	-	+	-	-	+	-	-	-	278.2	5.63
8	3	+	+	-	+	-	-	+	-	-	-	+	2390.0	7.78
9	12	+	-	+	-	-	+	-	-	-	+	+	16.0	2.77
10	2	-	+	-	-	+	-	-	-	+	+	+	4474.1	8.41
11	7	+	+	-	+	-	-	-	+	+	+	-	39.1	3.67
12	9	+		+	+	+	+	+	+	+	+	+	3196.0	8.07
$\Sigma XY =$		-1.90	6.46	-1.74	16.70	19.12	-0.80	-4.08	-10.22	3.82	-8.44	7.44		s = 1.98
$\frac{\Sigma XY}{T/2} =$		-0.32	1.08	-0.29	2.78	3.19	-0.13	-0.68	- 1.70	0.64	-1.41	1.24		t = 3.18
$\frac{\Sigma XY}{T/2}/2 =$		-0.16	0.54	-0.15	1.39	1.59	-0.07	-0.34	- 0.85	0.32	-0.70	0.62		b ± 1.82

FIGURE 11 - Cobalt-60 main effects design (IRC-718).

T Trial	Run Order	X_1 [Ni^{++}]	X_2 [Ca^{++}]	X_3 [Mg^{++}]	X_4 [OH^-]	X_5 [NH_4^+]	X_6 [$Co_3^=$]	X_7 [$PO_4^{\equiv}$]	X_8 [$SO_3^=$]	X_9	X_{10}	X_{11}	Y c/m/ml Eff	ln Counts
1	4	-	-	+	-	-	-	+	+	+	-	+	9.0	2.20
2	11	-	+	-	-	-	+	+	+	-	+	-	13.6	2.61
3	8	+	-	-	-	+	+	+	-	+	-	-	3.9	1.36
4	10	-	-	-	+	+	+	-	+	-	-	+	9206.6	9.13
5	5	-	-	+	+	+	-	+	-	-	+	-	49.4	3.90
6	6	-	+	+	+	-	+	-	-	+	-	-	128.2	4.85
7	1	+	+	+	-	+	-	-	+	-	-	-	179.3	5.19
8	3	+	+	-	+	-	-	+	-	-	-	+	47.5	3.86
9	12	+	-	+	-	-	+	-	-	-	+	+	16.2	2.79
10	2	-	+	-	-	+	-	-	-	+	+	+	7.8	2.05
11	7	+	-	-	+	-	-	-	+	+	+	-	307.7	5.73
12	9	+	+	+	+	+	+	+	+	+	+	+	115.8	4.75
ΣXY =		-1.06	-1.80	-1.06	16.02	4.34	2.56	-11.06	10.80	-6.54	-4.76	1.14		s = 1.37
$\frac{\Sigma XY}{T/2}$ =		-0.18	-0.30	-0.18	2.67	0.72	0.43	- 1.84	1.80	-1.09	-0.79	0.19		t = 3.18
$\frac{\Sigma XY}{T/2}/2$ =		-0.09	-0.15	-0.09	1.34	0.36	0.21	- 0.92	0.90	-0.55	-0.40	0.10		b ± 1.26

FIGURE 12 - Cobalt-60 main effects design (HCR-2W-H).

Figure 10 is the analysis for MSC-1. The confidence limits for the b's are ± 0.74. This analysis indicates [$SO_3^=$], [NH_4^+], and [OH^-] are all factors. Again, in comparison with Figure 6, [$CO_3^=$] is decreased significantly. Figure 11 is the analysis of IRC-718. This analysis indicates that none of the factors produced an effect, but does give higher values for [NH_4^+], [OH^-], and [$SO_3^=$] than any of the other factors. The erratic behavior of IRC-718 in comparison to the other cation exchangers is not fully understood. Figure 12 is the analysis of the data for HCR-2W-H. The confidence limits for the b's are ± 1.26. The analysis indicates [OH^-] is a factor and gives high values for [$SO_3^=$] and [$PO_4^{\equiv}$]. The low values for [NH_4^+] indicate that it might be the factor, referred to earlier, that was filtered from run 9 in the feed solution for HCR-2W-H.

At this point in the experiment, the results were reviewed in order to decide what factors were causing the large effect in run 10. The ln counts analysis indicated that [$CO_3^=$] is probably not involved. Since it is well known that cobalt carbonate complexes are unstable in aqueous solution [6,7,10-14] and, therefore, probably would not form under such conditions, a run which duplicated run 10 but contained no $CO_3^=$ was made using MSC-1. The result was that 28.00% of the cobalt was removed, compared to 28.98% removal in run 10 with $CO_3^=$. This difference is less than 1% and, therefore, is considered to be the same result. The ammonium and sulfite ions, however, are known to form negative complexes [8,9,16] such as [$Co(NH_3)_4(SO_3)_2$]$^-$with cobalt (III), and many cobalt complexes are pH dependent [14]. Also, many cobalt (III) complexes containing OH^- are known to exist [8-10].

Therefore, the factors chosen for the cobalt-60 interaction design were $[OH^-]$, $[NH_4^+]$, and $[SO_3^=]$.

DISCUSSION

Cobalt exists in various valence states; the most common of which are +2 and +3. The first three ionization potentials of cobalt [11] are 7.86, 17.05, and 33.49 eV. The high third ionization potential indicates that cobalt (III) compounds would be rare. In truth, the simple cobalt (III) compounds do not exist in aqueous solution and are, therefore, somewhat few in number [7]. Simple cobalt compounds found in aqueous solution will be in the cobalt (II) form. However, a vast number of cobalt (III) complex compounds exist and are generally very stable in aqueous solutions [7,8].

In contrast, cobalt (II) complexes are few and unstable [8]. In the solutions used in this experiment, all the cobalt was probably either in the form of cobalt (II) simple compounds or cobalt (III) complexes. Because the cobalt in cobalt (II) simple compounds would always be a dipositive cation, removal by the cation exchange resin would be easily accomplished. Cobalt (III) complexes, on the other hand, may have a positive, negative, or neutral charge. Cobalt (III) has a coordination number of six and primarily forms octahedral complexes [7,8,10,11]. The cations, except for NH_4^+, used in this experiment would not form negative or neutral complexes with cobalt and produced no effect through competition for resin sites. The ammonium ion, however, releases NH_3, according to the proton transfer reaction constant (pk = 9.245) [15].

For the reaction

$$NH_4^+ \rightleftarrows NH_3 + H^+ \qquad \text{(I)}$$

$$K = \frac{[NH_3]\ [H^+]}{[NH_4^+]}$$

$$pK = -\log K \quad \therefore K = 5.69 \times 10^{-10}$$

$[NH_4^+]$ added to solution = 50 ppm

$= 2.77 \times 10^{-3} M$

In reaction I at equilibrium, $[NH_3] = [H^+]$, and $[NH_4^+] = 2.77 \times 10^{-3} M - [NH_3]$

$$\therefore K = \frac{[NH_3]^2}{2.77 \times 10^{-3} M - [NH_3]} = 5.69 \times 10^{-10}$$

(NH_4^+ added to pH = 7 water)

$$[NH_3]^2 + 5.69 \times 10^{-10} [NH_3] - 1.58 \times 10^{-12} M = 0$$

$[NH_3] = 1.26 \times 10^{-6} M$ and $[NH_4^+] = 2.77 \times 10^{-3} M$

pH = 5.90

As base is added to attain pH = 10 or $[H^+] = 10^{-10}$, reaction (I) will be pulled to the right by reaction (II):

$$H^+ + OH^- \rightleftarrows H_2O \qquad \text{(II)}$$

At ph = 10,

$$5.69 \times 10^{-10} = \frac{[NH_3]\ 10^{-10}}{2.77 \times 10^{-3} M - [NH_3]}$$

$[NH_3] = 2.36 \times 10^{-3} M = 43$ ppm

$[NH_4^+] = 4.08 \times 10^{-4} M = 7$ ppm

At pH = 10, free NH_3 is present which is known to form many complexes with cobalt. [6-14]. Aqueous ammino cobalt (III) complexes are of the form $(Co(NH_3)_6]^{+3}$, $[Co(NH_3)_5H_2O]^{+3}$, $[Co(NH_3)_4(H_2O)_2]^{+3}$, etc. Since NH_3 is neutral, although it occupies up to six coordination positions on the cobalt atom, the complex is still positive. The complex can be neutral or negative only when cobalt is complexed with negatively charged ions. With hydroxide and sulfite ions, many such possible complexes exist. Some of these are $[CO(NH_3)_4(SO_3)_2]^-$, $[CO(NH_3)_3(SO_3)_3]^{-3}$, and $[Co(NH_3)_4(SO_3)(OH)]^\circ$ [8,9,16].

Since water can also coordinate with cobalt (III), the possibilities are numerous.

Cobalt interaction design experiment

The concentrations and variables for the cobalt interaction design are given in Table 14.

Table 14 - COBALT INTERATION DESIGN - VARIABLE CONCENTRATION

Ion	Level	Units	Value
NH_4^+	+	ppm	100
NH_4^+	0	ppm	50
NH_4^+	-	ppm	1
$SO_3^=$	+	ppm	100
$SO_3^=$	0	ppm	50
$SO_3^=$	-	ppm	1
OH^-	+	pH	10
OH^-	0	pH	6.5
OH^-	-	pH	3

The resins to be used in the cobalt-60 are: AG50WX8, MSC-1, HCR-2WH. All resins are strong acid cationic in sodium form. The constants are: $[^{60}Co]$ = 10,000 counts/min/ml and flow rate = 400 ml/min. The experimental design is given in Figure 13.

Summary of cobalt experiments

A main effects design experiment performed on cobalt-60 using eight variables and four ion exchange resins showed a large interaction of two or more variables which made single variable effects difficult to determine. Variables picked for inclusion in the interaction effects design were $[OH^-]$, $[NH_4^+]$, and $[SO_3^=]$. The plan for the interaction design was outlined.

Iodine main effects design experiment

A main effects experimental design was prepared to study factors that could possibly affect the adsorption of iodine-129 on anion exchange resins. Since the conditions necessary for formation of the I^+ ion do not normally occur in dilute waste water solutions, the possibility of positive iodine ions being present was disregarded. Other inorganic complexes that I^- would form, such as I_3^-, would normally be negative complexes. Since iodine is normally in a negative ionic state in aqueous solutions, and the complexes considered most likely to be present are negatively charged also, only negatively charged "competition" type ions were chosen

Trial	Order	$[OH^-]$	$[NH_4^+]$	$[SO_3^=]$	x_1x_2	X_1X_3	X_2X_3	Y's	$\bar{Y}$	$(Y-\bar{Y})$	$(Y-\bar{Y})^2$	S^2	df r-1	y_{prd}	$y_{pred.} - y_{obs.}$
CP	1	o	o	o	o	o	o								
CP	10	o	o	o	o	o	o								
CP	19	o	o	o	o	o	o								
1	5 16	-	-	-	+	+	+								
2	6 14	+	-	-	-	-	+								
3	7 11	-	+	-	-	+	-								
4	4 17	+	+	-	+	-	-								
5	8 13	-	-	+	+	-	-								
6	3 15	+	-	+	-	+	-								
7	2 18	-	+	+	-	-	+								
8	9 12	+	+	+	+	+	+								

$\Sigma X\bar{Y}$ =

$\Sigma XY/(T/2)$ =

$\frac{2\Sigma\ x\ \ y/T}{2} = b =$

FIGURE 13 - Cobalt-60 interaction design

as variables. The variablesand their levels are presented in Table 15. The constants used were: Flow rate = 0.5 ± 0.1 bed volumes/min and $[I^{129}]$ = 100 counts/min/ml. The resins used were IRA-938, IRA-430, MSA-1, and SAR, all in chloride form. A copy of the main effects experimental design is presented in Figure 14.

In ion exchange experiments, columns as large as the engineering colums are generally used to determine hydraulic operational factors and to study long-term effects of the ion exchange material [17]. For studies of chemical factors only, much smaller columns are generally used [17]. The resin bed volume used in this experiment is 1/5 of that of the engeineering columns. The columns used in this experiment are large enough to study hydraulic scale-up factors, [17] but these factors have already been studied on the engineering columns for resins of the size and density used in this experiment.

The intent of this experiment is only to determine the chemical factors that affect the removal of iodine-129 from aqueous solution by anion exchange resins.

Since a column as large as the engineering columns is not necessary for this experiment, the smaller columns were used. Using the smaller columns allowed the iodine main effects experiment to be performed while the cobalt main effects experiment was being done on the engineering columns.

Table 15 - VARIABLES AND THEIR LEVELS

Variable	X_1	X_2	X_3	X_4	X_5	X_6
Variable Name	$[Cl^-]$	$[PO_4^{\equiv}]$	$[SO_3^=]$	$[OH^-]$	$[SO_4^=]$	$[CO_3^=]$
(+) Level	1500 ppm	700 ppm	10 ppm	pH = 10	10 ppm	1000 ppm
- Level	R.O.W.[a]	R.O.W.[a]	R.O.W.[a]	pH = 3	R.O.W.[a]	R.O.W.[a]
M (+) Level (moles/liter)	0.0423	0.0074	1.25×10^{-4}	10^{-4}	1.04×10^{-4}	0.0167

[a] Reverse osmosis water.

T Trial	Run Order	X_1 $[Cl^-]$	X_2 $[PO_4^{\equiv}]$	X_3 $[SO_3^=]$	X_4 $[OH^-]$	X_5 $[SO_4^=]$	X_6 $[CO_3^=]$	X_7	X_8	X_9	X_{10}	X_{11}	Y % I^{129} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	
2	11	-	+	-	-	-	+	+	+	-	+	-	
3	8	+	-	-	-	+	+	+	-	+	-	-	
4	10	-	-	-	+	+	+	-	+	-	-	+	
5	5	-	-	+	+	+	-	+	-	-	+	-	
6	6	-	+	+	+	-	+	-	-	+	-	-	
7	1	+	+	+	-	+	-	-	+	-	-	-	
8	3	+	+	-	+	-	-	+	-	-	-	+	
9	12	+	-	+	-	-	+	-	-	-	+	+	
10	2	-	+	-	-	+	-	-	-	+	+	+	
11	7	+	-	-	+	-	-	-	+	+	+	-	
12	9	+	+	+	+	+	+	+	+	+	+	+	
$\Sigma XY =$													
$\frac{\Sigma XY}{T/2} =$													
$\frac{2\Sigma XY/T}{2} =$													

FIGURE 14 - Iodine-129 main effects experimental design

PROCEDURE

The procedure used in this experiment was first to prepare the feed solution for each run according to the experimental design (Figure 14), the variable concentrations (Table 15), and sample. Then each solution was run through the ion exchangers, and three samples were taken evenly over the course of the run. The samples were then prepared for counting and counted in a Packard 460 CD scintillation counter. The resins were regenerated with a solution of 1.0 N HCl with 1.0 N KCl until the regenerant counts were reduced to background, and were then washed with reverse osmosis water for approximately 10 bed volumes (2 liters).

RESULTS

Figures 15 through 18 show the main effects design data analyses of resins IRA 938, IRA 430, MSA-1, and SAR respectively. Figure 15, the analysis for IRA 938, has b confidence limits of ± 1.59. This value places zero in the range of the b's for all the variables and, therefore, indicates no variable effects. The average y value for all the runs is 98.61%. Figure 16, the data analysis for IRA 430, has b confidence limits of ± 0.61. This value places zero in the range of the b's for all the variables and, therefore, indicates no variable effects. The average y value for all the runs is 98.89%. Figure 17, the data analysis for MSA-1, has b confidence limits of ± 1.11. This value places zero in the range of the b's for all the variables and, therefore, indicates no variable effects. The average value of the y's for all the runs is 98.47%. Figure 18, the data analysis for SAR, has b confidence limits of ± 0.85. This value places zero in the range of the b's for all the variables and, therefore, indicates no variable effects. The average value of the y's for all the runs is 98.44%. The average iodine concentration of all 144 effluent samples in the experiment was 1.45 counts/min/ml.

DISCUSSION

Iodine with its [Kr] $4d^{10}$ $5s^2$ $5p^5$ electron structure exhibits the halogen tendency to form the I^- ion in combination with metals. However, being the largest of the stable halogens, the nucleus is much more effectively screened which makes iodine the most electropositive of the common halogens [18]. Because of its relatively low electron affinity, iodine produces the positive valences +1, +3, +5, +7 much more readily than do the other common halogens [18]. Iodine also forms positive complex ions such as IO^+, IO_2^+, IO_3^+ and reacts with the other halogens forming compounds such as IBr_3, ICl, and IF_7, in which it has positive valences.

Since the negative ions introduced as factors in the solutions for this experiment produced no effects, it is believed that no positive valence iodine was present. No positive valences of iodine were expected but had they occurred they would have produced a large negative effect since they would not be removed by anion exchange resins. Iodine forms bonds that have a high covalent nature [18], and it is possible that this covalent nature could have impaired the effectiveness of the ion exchange resins. This partial covalent nature could in fact be the reason that only 98% of the iodine was removed. An interaction design involving any of

T Trial	Run Order	X_1 $[Cl^-]$	X_2 $[PO_4^{\equiv}]$	X_3 $[SO_3^{=}]$	X_4 $[OH^-]$	X_5 $[SO_4^{=}]$	X_6 $[CO_3^{=}]$	X_7	X_8	X_9	X_{10}	X_{11}	Y % I^{129} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	99.52
2	11	-	+	-	-	-	+	+	+	-	+	-	99.71
3	8	+	-	-	-	+	+	+	-	+	-	-	99.22
4	10	-	-	-	+	+	+	-	+	-	-	+	92.69
5	5	-	-	+	+	+	-	+	-	-	+	-	99.33
6	6	-	+	+	+	-	+	-	-	+	-	-	99.93
7	1	+	+	+	-	+	-	-	+	-	-	-	97.95
8	3	+	+	-	+	-	-	+	-	-	-	+	98.43
9	12	+	-	+	-	-	+	-	-	-	+	+	99.87
10	2	-	+	-	-	+	-	-	-	+	+	+	98.56
11	7	+	-	-	+	-	-	-	+	+	+	-	99.24
12	9	+	+	+	+	+	+	+	+	+	+	+	98.89
ΣXY =		3.86	3.60	7.64	-6.32	-10.06	-2.72	6.86	-7.34	7.38	7.86	-7.42	
$\frac{\Sigma XY}{T/2}$ =		0.64	0.60	1.27	-1.05	- 1.68	-0.45	1.14	-1.22	1.23	1.31	-1.24	
$\frac{2\Sigma XY/T}{2}$ =		0.32	0.30	0.64	-0.53	- 0.84	-0.23	0.57	-0.61	0.62	0.66	-0.62	

s = 2.14 5df

t = 2.57 95%

b ± 1.59

$\bar{y}$ = 98.61

FIGURE 15 - Iodine-129 main effects design (Resin: IRA 938)

T Trial	Run Order	X_1 $[Cl^-]$	X_2 $[PO_4^{\equiv}]$	X_3 $[SO_3^{=}]$	X_4 $[OH^-]$	X_5 $[SO_4^{=}]$	X_6 $[CO_3^{=}]$	X_7	X_8	X_9	X_{10}	X_{11}	Y % I^{129} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	99.13
2	11	-	+	-	-	-	+	+	+	-	+	-	99.09
3	8	+	-	-	-	+	+	+	-	+	-	-	99.11
4	10	-	-	-	+	+	+	-	+	-	-	+	97.86
5	5	-	-	+	+	+	-	+	-	-	+	-	99.71
6	6	-	+	+	+	-	+	-	-	+	-	-	99.81
7	1	+	+	+	-	+	-	-	+	-	-	-	97.46
8	3	+	+	-	+	-	-	+	-	-	-	+	97.94
9	12	+	-	+	-	-	+	-	-	-	+	+	99.77
10	2	-	+	-	-	+	-	-	-	+	+	+	98.05
11	7	+	-	-	+	-	-	-	+	+	+	-	98.94
12	9	+	+	+	+	+	+	+	+	+	+	+	99.76
ΣXY =		-0.67	-2.41	4.65	1.41	-2.73	4.17	2.85	-2.15	2.97	4.01	-1.61	
$\frac{\Sigma XY}{T/2}$ =		-0.11	-0.40	0.78	0.24	-0.46	0.70	0.48	-0.36	0.50	0.67	-0.27	
$\frac{2\Sigma XY/T}{2}$ =		-0.06	-0.20	0.39	0.12	-0.23	0.35	0.24	-0.18	0.25	0.33	-0.13	

s = 0.82 5df

t = 2.57 95%

b ± 0.61

$\bar{y}$ = 98.89

FIGURE 16 - Iodine-129 main effects design (Resin: IRA 430).

T Trial	Run Order	X_1 $[Cl^-]$	X_2 $[PO_4^{\equiv}]$	X_3 $[SO_3^{=}]$	X_4 $[OH^-]$	X_5 $[SO_4^{=}]$	X_6 $[CO_3^{=}]$	X_7	X_8	X_9	X_{10}	X_{11}	Y % I^{129} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	99.33
2	11	-	+	-	-	-	+	+	+	-	+	-	99.26
3	8	+	-	-	-	+	+	+	-	+	-	-	98.63
4	10	-	-	-	+	+	+	-	+	-	-	+	95.66
5	5	-	-	+	+	+	-	+	-	-	+	-	99.43
6	6	-	+	+	+	-	+	-	-	+	-	-	99.63
7	1	+	+	+	-	+	-	-	+	-	-	-	97.87
8	3	+	+	-	+	-	-	+	-	-	-	+	95.92
9	12	+	-	+	-	-	+	-	-	-	+	+	99.52
10	2	-	+	-	-	+	-	-	-	+	+	+	98.29
11	7	+	-	-	+	-	-	-	+	+	+	-	98.71
12	9	+	+	+	+	+	+	+	+	+	+	+	99.41
ΣXY =		-1.52	-0.92	8.74	-4.12	-3.10	2.58	2.32	-1.16	6.32	7.56	-5.42	
$\frac{\Sigma XY}{T/2}$ =		-0.25	-0.15	1.46	-0.69	-0.52	0.43	0.39	-0.19	1.05	1.26	-0.90	
$\frac{2\Sigma XY/T}{2}$ =		-0.13	-0.08	0.73	-0.34	-0.26	0.22	0.19	-0.10	0.53	0.63	-0.45	

s = 1.49 5df

t = 2.57 95%

b ± 1.11

$\bar{y}$ = 98.47

FIGURE 17 - Iodine-129 main effects design (Resin: MSA-1).

T Trial	Run Order	X_1 $[Cl^-]$	X_2 $[PO_4^{\equiv}]$	X_3 $[SO_3^{=}]$	X_4 $[OH^-]$	X_5 $[SO_4^{=}]$	X_6 $[CO_3^{=}]$	X_7	X_8	X_9	X_{10}	X_{11}	Y % I^{129} Removed
1	4	-	-	+	-	-	-	+	+	+	-	+	98.77
2	11	-	+	-	-	-	+	+	+	-	+	-	99.13
3	8	+	-	-	-	+	+	+	-	+	-	-	98.43
4	10	-	-	-	+	+	+	-	+	-	-	+	96.93
5	5	-	-	+	+	+	-	+	-	-	+	-	99.34
6	6	-	+	+	+	-	+	-	-	+	-	-	99.43
7	1	+	+	+	-	+	-	-	+	-	-	-	97.92
8	3	+	+	-	+	-	-	+	-	-	-	+	96.21
9	12	+	-	+	-	-	+	-	-	-	+	+	99.39
10	2	-	+	-	-	+	-	-	-	+	+	+	97.42
11	7	+	-	-	+	-	-	-	+	+	+	-	99.06
12	9	+	+	+	+	+	+	+	+	+	+	+	99.23
ΣXY =		-0.28	-2.58	6.90	-0.86	-3.02	3.82	0.96	0.82	3.42	5.88	-5.36	
$\frac{\Sigma XY}{T/2}$ =		-0.05	-0.43	1.15	-0.14	-0.59	0.64	0.16	0.14	0.57	0.98	-0.89	
$\frac{2\Sigma XY/T}{2}$ =		-0.02	-0.22	0.50	-0.07	-0.25	0.32	0.08	0.07	0.29	0.49	-0.45	

s = 1.14 5df

t = 2.57 95%

b ± 0.85

$\bar{y}$ = 98.44

FIGURE 18 - Iodine-129 main effects design (Resin: SAR).

these variables is not necessary because none of the factors affected the removal of the iodine. If an interaction design were performed, it would probably produce a prediction equation similar to y = 98%.

SUMMARY

A main effects design for iodine-129 was performed using [I], $[PO_4^{=}]$, $[SO_3^{=}]$, $[OH^-]$, $[SO_4^{=}]$, and $[CO_3^{=}]$ as factors. The resins used were al anion exchange resins in chloride form. One of the factors affected the removal of iodine-129 from aqueous solution.

Treatment of Cesium Contaminated Liquid Waste

William H. Bond

Introduction

The Waste Disposal group at Mound Facility is responsible for processing low-level contaminated aqueous wastes generated during normal operations. Predominately these wastes are contaminated only with plutonium-238. Occasionally, other isotopes occur in the waste which require minor process alterations. Usually, these isotopes are actinides and are easily removed. In one instance, however, cesium-137 was the major contaminant in a 4,600-gal batch.

Cesium cannot be removed by the standard process (pH adjustment followed by flocculation), so a cesium removal step had to be added to the standard process.

An alternative to processing was to mix the waste with cement in 55-gal drums and send it to shallow land burial. Although this option possessed several attractive features, the cost was rather high ($37,000). Because of this high cost and the large number of drums that would be sent for burial, a cesium removal process that would permit the waste to be normally processed, and so, avoid burying such a large quantity of water, appeared to be economically attractive. The cost of drumming the waste was, therefore, used as the upper limit for any prospective cesium removal process. Also, the chosen process was required to meet radiological safety standards during processing and to possess an immobilizable final waste form, a reasonably short processing time, short overall project duration, and as high as possible cesium removal.

The characteristics of the known cesium removal processes were evaluated against these criteria and in consideration of the fact that this waste, which was at pH of 12.8 and contained 176 kg of sodium (11 g/liter), was not likely to be encountered again. The physical processes, such as reverse osmosis and evaporation, were eliminated because equipment costs could not be amortized, thus making the processsing cost excessive. Organic ion exchange resins in both hydrogen and sodium forms were considered as one-time adsorbents as well as recycled ion exchangers. Because of the high sodium concentration in the waste, both the sodium and hydrogen form organic resins failed to show appreciable benefit over drumming and burying. Inorganic adsorbents, primarily zeolites, were found to be rather attractive in cost and final volume when mixed with the waste and allowed to settle. Drago and Buchholz, however, achieved a maximum of 64% cesium removal with clinoptilolite in a solution containing less sodium than the waste under consideration [19]. Clinoptilolite, when placed in columns, increases cesium removal, but the use of columns increases process cost and residual solid waste volume. Although normally incidental to the choice of a separation process, column availability and manpower for fabricating and piping the column array were major deterants to the use of ion exchange columns. Therefore, neither organic nor inorganic adsorbents were considered to be optimum treatments for the waste under the existing conditions.

Chemical processes, primarily precipitation, were attractive because equipment requirements for these processes closely matched the equipment that was on hand. Two processes received the most attention: ferrocyanide extraction and sodium tetraphenyl boron coprecipitation. Ferrocyanide processes for cesium have been fairly well documented, [20-22] but although sodium tetraphenyl boron is well known in the analytical laboratory, its use on a large scale is rare. This limited use of sodium tetraphenyl boron for large volumes of waste has likely been because of its cost, which is quite high through regular supply houses. A custom manufacturer* of small quantities (a few kilograms to a few thousand kolograms) of complex chemicals has been found, however, who will supply sodium tetraphenyl boron, in relatively large quantities, for about 20% of the usual cost. This price makes sodium tetraphenyl boron competitive with one-time organic resins.

Descriptions of the development of the ferrycyanide and sodium tetraphenyl boron processes, treatment, and results were included in the previous report [3].

Processing the waste

Eight kilograms of sodium tetraphenyl boron were dissolved in 32 liters of distilled water and then diluted to 40 gal. This solution was added to the 4,600 gal of waste over a 3-hr period to the outlet of a 30 gal/min pump set up to transfer the waste from the outer section of the flocculator to the inner. This method provided good mixing of the sodium tetraphenyl boron with the waste because the 4,600 gal of waste should have been recycled in about 2.5 hr.

Subsequent to mixing, a sample of the waste was taken and assayed for cesium. The cesium concentration was 16 counts/min/ml (7.2 pCi/ml). The waste was then sampled for potassium. Residual potassium was found, which raised the concern that the cesium that was already precipitated might be exchanged for the potassium remaining in the waste. In an attempt to prevent this from occurring, an additional 1.5 kg of sodium tetraphenyl boron was dissolved in 8 liters of distilled water, diluted to 10 liters, and added to the clariflocculator over a 2 hr period as described previously. The final concentration of sodium tetraphenyl boron used for the process was 545 mg/liter of waste. Cesium concentration of the filtered supernatant resulting from the sodium tetraphenyl boron treatment was 4.0 counts/min/ml ($\sim$1.81 ± 10% pCi/ml).

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The mixture resulting from the treatment was allowed to settle, and the supernatant was pumped to another clariflocculator to separate the sludge and supernatant portions in the event that cesium might be released from the settled sludge and recontaminate the supernatant. This precaution was found to be unnecessary, because the small amount of supernatant remaining on the sludge was assayed several weeks after treatment and found to have the same cesium concentration as the removed supernatant. Therefore, the precipitate was found to be stable in the presence of an excess of potassium in the supernatant.

The transferred supernatant was processed by the standard Waste Disposal treatment and discarded from the plant. The precipitate and sludge were mixed with cement in 55-gal drums and shipped for shallow land burial.

Summary

A process for the coprecipitation of cesium-137 with potassium, using sodium tetraphenyl boron, from a salty low-level, aqueous waste was developed and successfully conducted. The cesium-137 concentration in the supernatant of the waste was reduced from 570 to 4 counts/min/ml.

Because the process is coprecipitation, the presence of a small amount of potassium is required for low levels of cesium. A sufficiently high level of cesium would require no potassium for precipitation. However, the addition of "NALCO 676" initiated cesium precipitation in the absence of potassium during the non-mixing test. Moderate solution mixing, or the addition of "NALCO 676", is required for coprecipitation to occur.

Sodium tetraphenyl boron, in bulk quantities, is economically competitive with physical processes under certain conditions. (See Table 16.) A savings of $33,368 was realized by processing the waste with sodium tetraphenyl boron instead of drumming.

Table 16 - PROCESS COST COMPARISON

Drumming cost		$37,000
Sodium tetraphenyl boron cost		
Material costs:		
Sodium tetraphenyl boron - 10 kg	$2500	
Drums, cement, shipping	152	
Labor cost:[a]		
Process water, drum sludge, etc.	980	
Total		$ 3,632

[a] Development costs not included.

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