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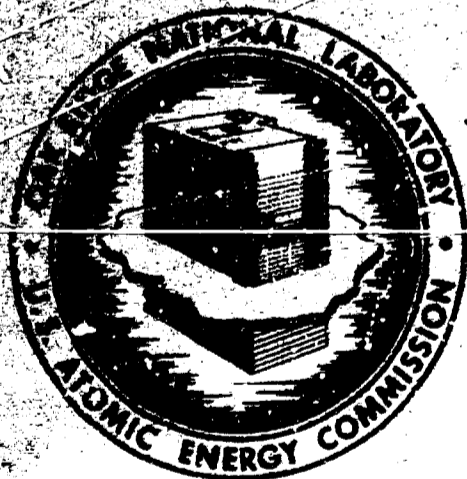
UC-70 - Waste Disposal and Processing

MASTER

**LOW-LEVEL RADIOACTIVE WASTE TREATMENT:
THE WATER RECYCLE PROCESS**

W. C. Yee
F. DeLora
W. E. Shockley

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CHEMICAL TECHNOLOGY DIVISION
Chemical Development Section B

LOW-LEVEL RADIOACTIVE WASTE TREATMENT: THE WATER RECYCLE PROCESS

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JULY 1970

*Guest scientist from the Spanish Atomic Energy Commission. Present Address:
Ministerio de Industria, Junta de Energia Nuclear, Madrid, Spain.

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LOW-LEVEL RADIOACTIVE WASTE TREATMENT: THE WATER RECYCLE PROCESS

W. C. Yee, F. DeLora,* and W. E. Shockley

ABSTRACT

The water recycle process was developed for decontaminating low-level radioactive aqueous wastes and recycling the purified water for reuse at nuclear installations. This process was successfully demonstrated through several cycles in a micro-pilot plant, using ORNL waste that contained low concentrations of salts and radionuclides. The recycle of water should be an improvement over present methods in which the wastes are decontaminated and the purified water is discharged to the environment. The steps in the process are: (1) clarification, using zeta-potential-controlled additions of coagulants such as alum, coagulant aids, activated silica, and nonionic organic polyelectrolyte; (2) demineralization by cation-anion exchange; and (3) treatment with activated carbon. Zeta-potential control was shown to be an excellent method for obtaining optimum clarification conditions. The water in the micro-pilot plant was decontaminated from all major radionuclides by factors (DF's) of 1000 to 10,000 for up to 2400 volumes of water per volume of cation resin. The DF's for cobalt (^{60}Co) and ruthenium (^{106}Ru) were 10^4 and 10^3 , the highest obtained in the ORNL waste development program.

1. INTRODUCTION

Low-level radioactive wastes, consisting of water with low concentrations of salts and radionuclides, are produced at most nuclear installations. Conventional decontamination methods^{1,2} include scavenging precipitation, ion exchange, or distillation, followed by discharge of the purified water to the environment. The objectives of the work reported here are: (1) to determine the technical feasibility of recycling the purified water for reuse (e.g., to determine the effects of the

*Guest scientist from the Spanish Atomic Energy Commission. Present address: Ministerio de Industria, Junta de Energia Nuclear, Madrid, Spain.

concentrations of contaminants in recycled water; and (2) to develop improved decontamination methods. Recycle appears desirable since it would eliminate the cost of purifying the water that is taken from rivers for use in the nuclear station; also, it would allow the nuclear station to become self-contained. Improved decontamination would, of course, decrease the flow of radionuclides to the environment. The radioactive concentrate could be incorporated in asphalt or polyethylene prior to disposal in a government burial ground.^{3,4}

Some of the solids in the waste are present in the form of colloidal particles. Thus, efficient clarification is important as the first treatment step. Zeta-potential control was developed as a regulatory method since previous workers had reported that it showed considerable promise.⁵

The second step is demineralization for efficient removal of soluble salts and radionuclides. This portion of the process differs from the previously developed process⁶⁻⁹ in which a high degree of decontamination, but only partial demineralization, was achieved. The third step is sorption on activated carbon. This sorption system was evaluated in an effort to increase the removal of radionuclides, such as ruthenium and cobalt, which had proved to be refractory in previous studies.

The water recycle process is compatible with present public opinion and with government policies that encourage the recirculation of waters and the minimization of discharge of both radioactive and nonradioactive salts to the environment.¹⁰ This type of process should also be applicable to the recycle of nonradioactive industrial and urban waste waters that require clarification and demineralization.

2. PROCESS FLOWSHEET

The water recycle process, which was developed for the treatment of low-level radioactive waste having a low salt content (Fig. 1), consists of the following steps: coagulation and clarification, demineralization, and treatment with activated carbon. The decontaminated and demineralized water is returned to the nuclear

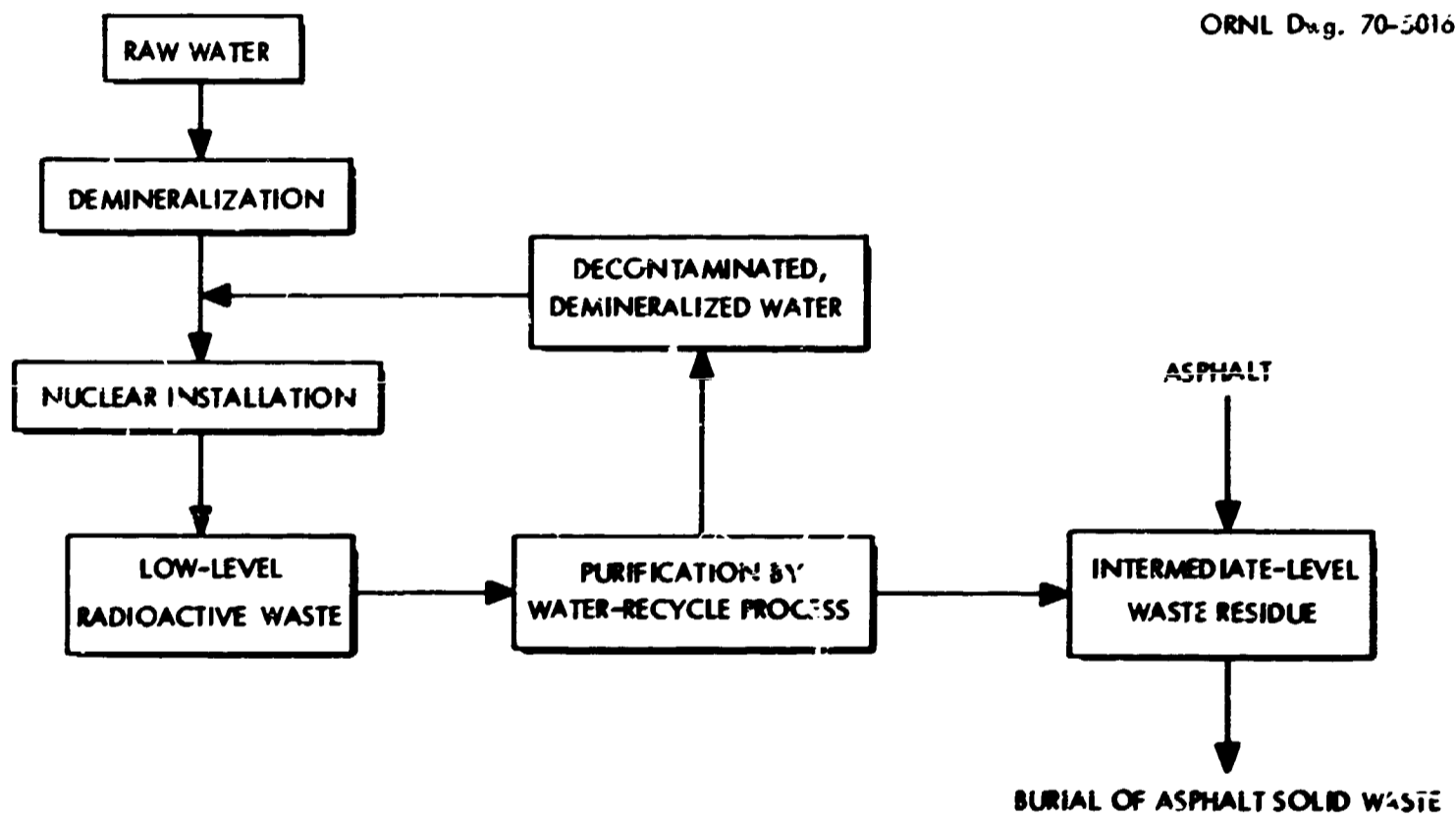


Fig. 1. Treatment of Low-Level Radioactive Waste and Disposal of Residues.

installation for reuse, while the concentrated radioactive waste is denitrated, if necessary, and then incorporated into asphalt and buried.

The flowsheet (Fig. 2) includes: (1) coagulation by zeta-potential-controlled additions of the coagulant, alum, and the coagulant aids; activated silica; and a nonionic organic polyelectrolyte; (2) clarification by upflow through a fluidized bed of sludge (upflow clarifier), followed by filtration through a bed of anthracite coal and sand; (3) demineralization by cation-anion exchange; and (4) sorption of the remaining radioactive and nonradioactive contaminants on granular activated carbon. Continuous operation of the coagulation-clarification system under optimum conditions is important to the maintenance of high decontamination factors (DF's) for radionuclides such as cobalt and ruthenium, which, in neutral wastes, exist in several forms including ionic and nonionic colloids. Efficient removal of these colloids depends on the neutralization of their mutually repelling surface charges, that is, their zeta-potential (ZP).⁵ The successive addition of activated silica (0.5 ppm as SiO₂) and alum [2 to 3 ppm as Al₂(SO₄)₃] in the flash mixer increased the ZP of synthetic recycle water from -20 mv to +3 mv. Stirring at 1750 rpm effectively enmeshed the colloids within the freshly formed aluminum hydroxide floc. The addition of 75 ppb of a nonionic organic polyelectrolyte* at the flocculator enhanced the growth and settling properties of the floc (Fig. 3) and accomplished the desired filtering action in the upflow clarifier (the primary filtering medium) by maintaining a fluidized bed of sludge. The final ZP was -2 mv. After filtration through anthracite and sand, the water had a clarity comparable to that of distilled water, representing a greater than 100-fold decrease in turbidity. The micro-pilot plant components used in these tests are shown in Figs. 4 and 5.

Demineralization was accomplished by ion exchange, using two separate columns; one contained 1.0 cation resin volume (400 ml) of a strong-acid cation resin (Dowex 50W-X8, 20 to 50 mesh), and the other contained 1.9 cation resin volumes (750 ml)

*Purifloc N-12, product of the Dow Chemical Co., Midland, Mich.

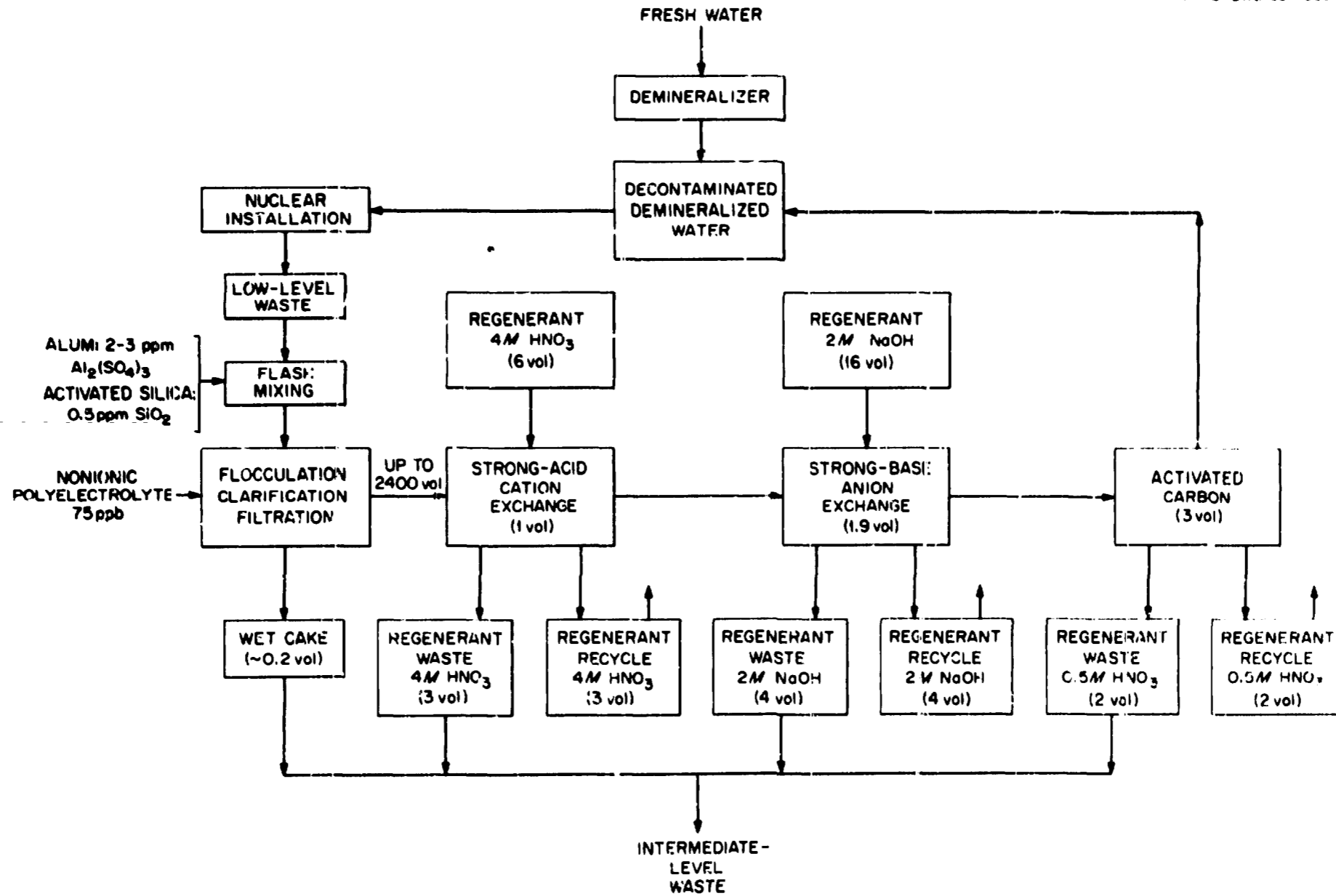


Fig. 2. Flowsheet for the Water Recycle Process.

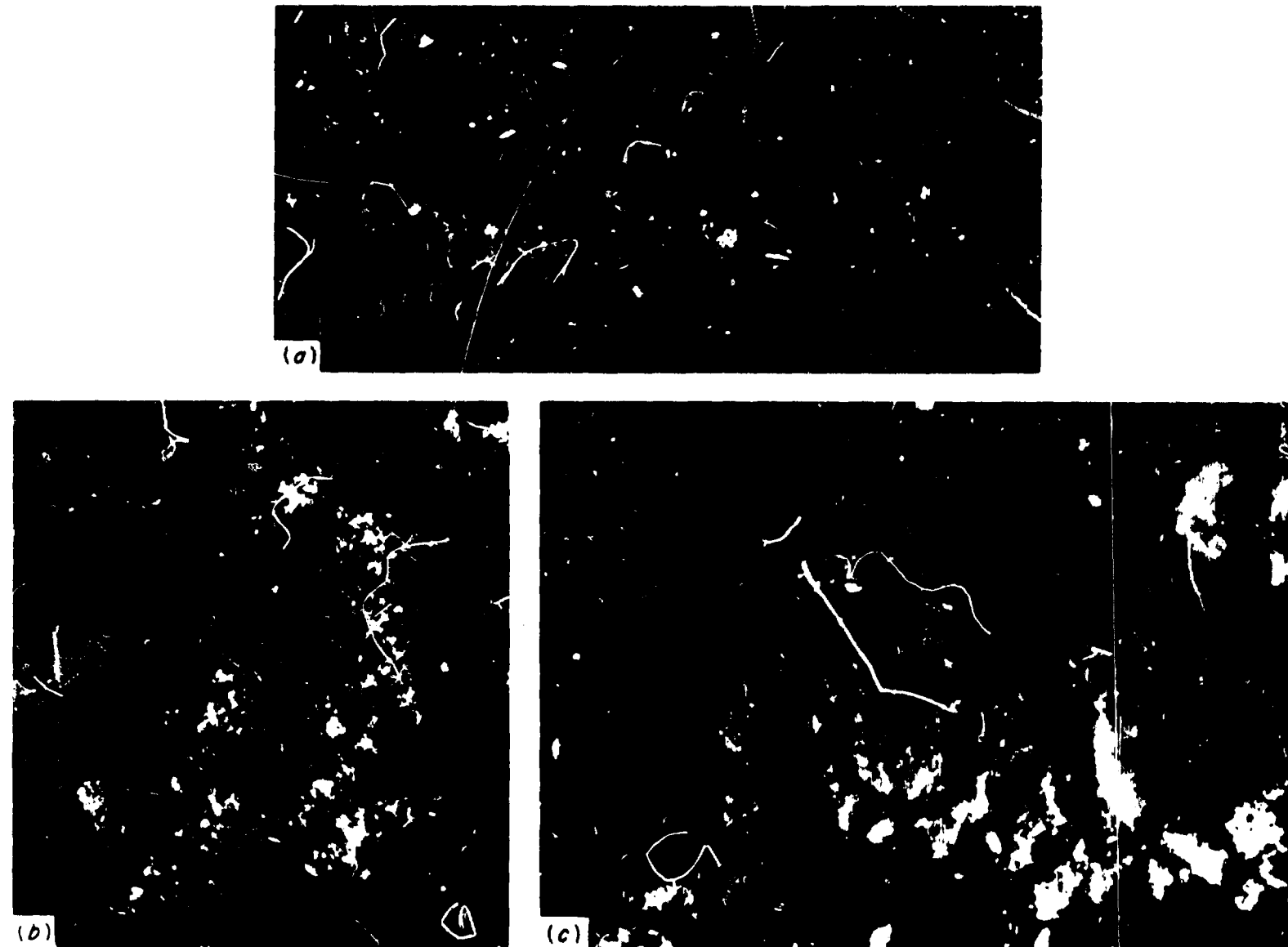


Fig. 3. Coagulation of Suspensoids by Use of Zeta-Potential Control. (a) Raw water contains suspended particles which have an average zeta potential (ZP) of -20 mv. (b) Alum [2 to 3 ppm $\text{Al}_2(\text{SO}_4)_3$] and activated silica (0.5 ppm, as SiO_2) are added with rapid stirring. Repulsive surface charges are neutralized to a ZP of $+3$ mv, and particles (the "stars") are enmeshed into the freshly formed small aggregates of aluminum hydroxide floc. (c) Floc growth is enhanced by the addition of a nonionic organic polyelectrolyte (75 ppb) which lowers the ZP to -2 mv. The water, after filtration, has a clarity comparable to that of distilled water.

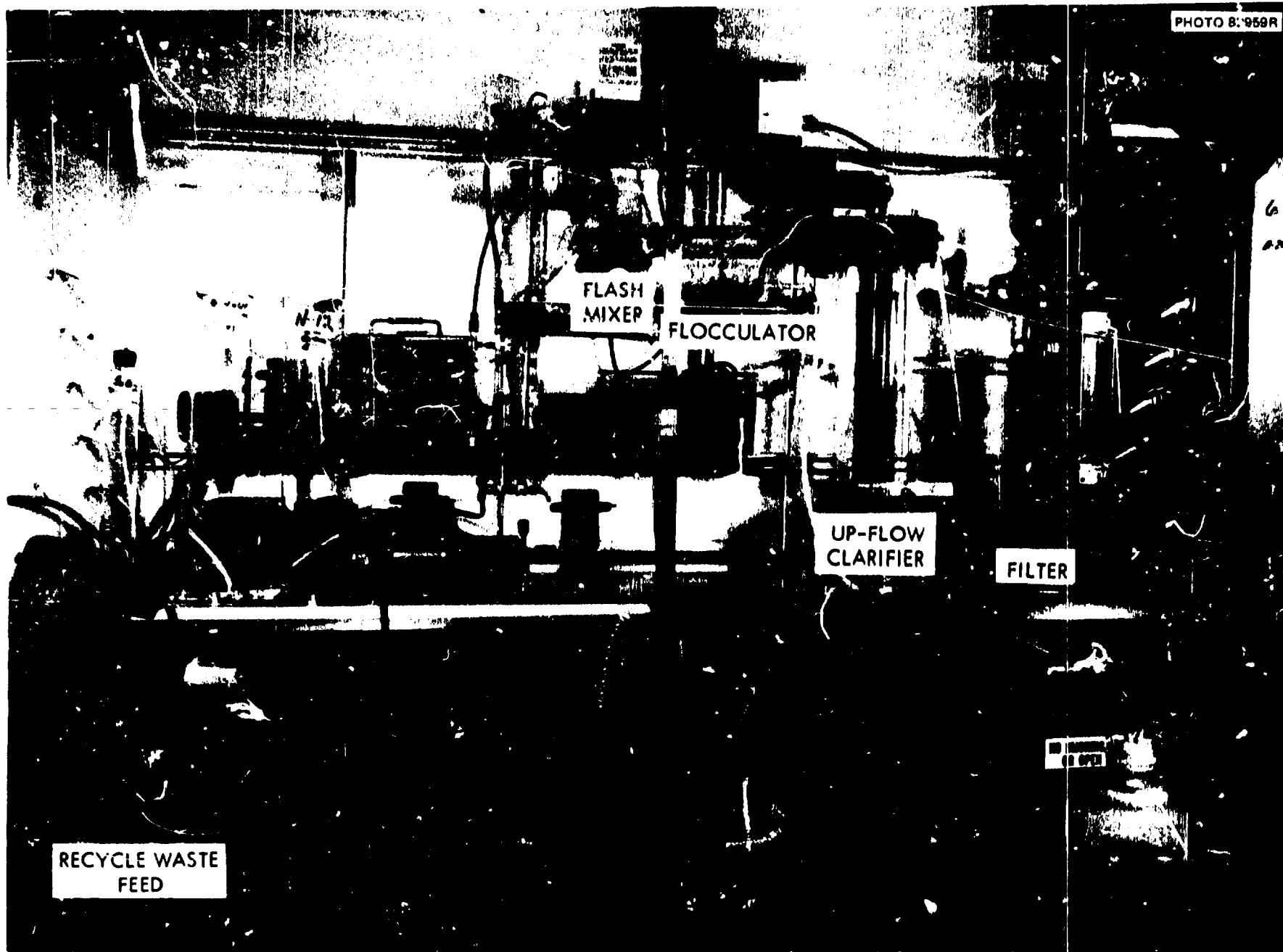


Fig. 4. Micro-Pilot Plant Used in Tests of the Water Recycle Process: Clarification Section.

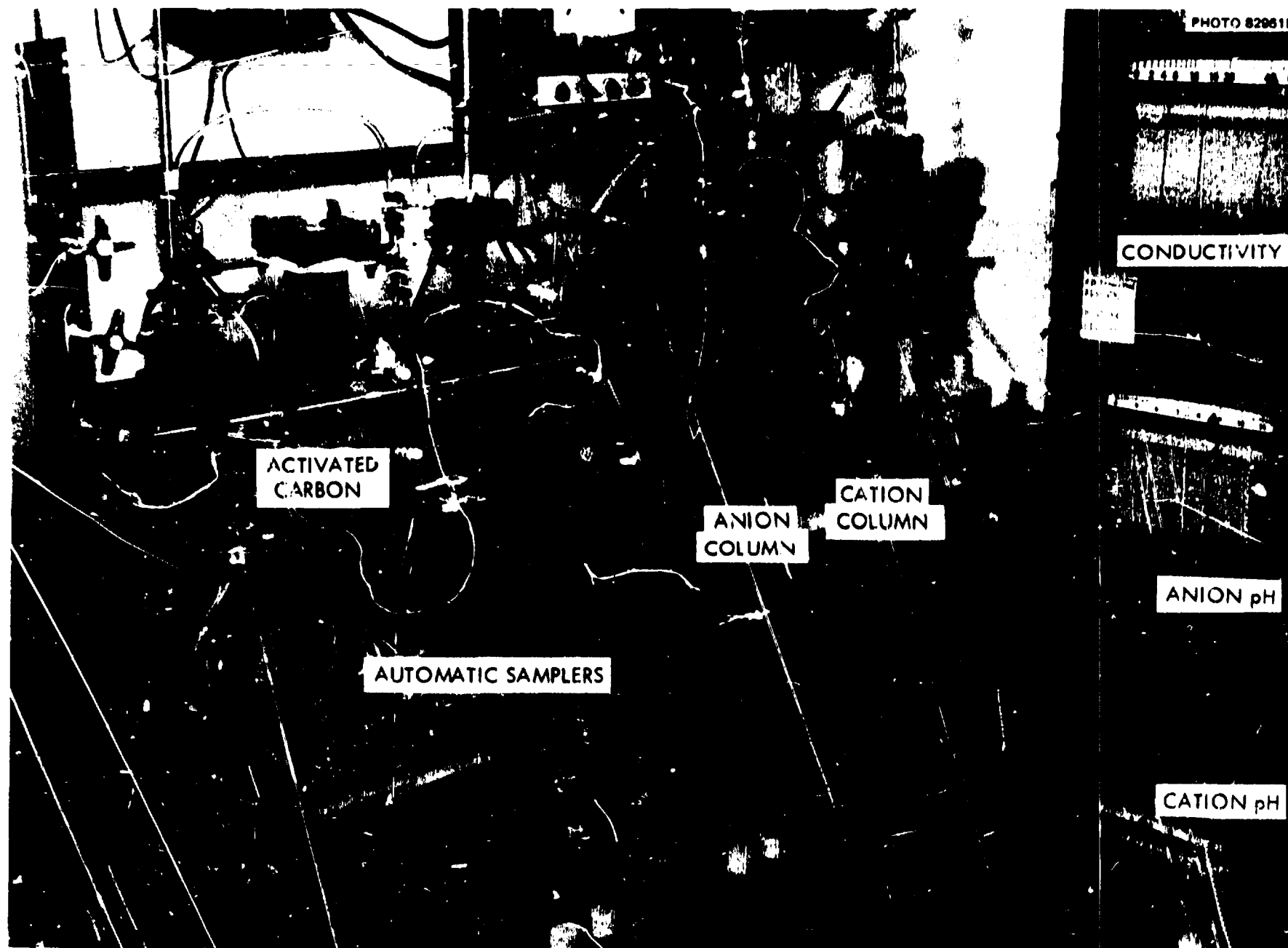


Fig. 5. Micro-Pilot Plant Used in Tests of the Water Recycle Process: Demineralization-Carbon Section.

of a strong-base anion resin (Dowex 1-X8, 20 to 50 mesh). The contact times for the cation and anion columns were 2.6 and 4.8 min, respectively. The flow rate was 150 ml/min. A third column, containing activated carbon, was used for final polishing (or cleaning). A vegetable-based granular, activated carbon* was used in preference to other commercial carbons because of its greater capacity for cobalt. The volume of the activated-carbon column (i.e., 1200 ml or 3.0 cation resin volumes) was maintained at three times that of the cation column to obtain the optimum residence time of 8 min recommended by Roberts and Abdel-Razek.¹¹

The water used in the micro-pilot plant evaluation of the flowsheet had to be similar to water that had been demineralized and then recontaminated through plant use. Consequently, the waste used in these experimental runs was water with a low solids content and consisted of: (1) 80%, by volume, of decontaminated and demineralized ORNL-LLW treated via the water recycle process, (2) 20% raw ORNL-LLW, and (3) radioactive tracers. The specific conductance of the composite waste water was 80 to 100 micromhos/cm, only one-fourth to one-third of that for ORNL-LLW. Equal amounts of tracer ^{106}Ru were added from two sources, namely, an acid solution containing ^{106}Ru as the pure isotope, and ORNL intermediate-level waste (pH, 12 to 13) containing ^{106}Ru . Cobalt-60 from these two sources was also present. Overall DF's of 100 to 10,000 for all major radioactive species were obtained for up to 2400 volumes of waste treated per bed volume of cation resin; DF's in the few-thousand range were achieved for ^{131}I , ^{106}Ru , ^{90}Sr , and total rare earths, while DF's of 14,000 and 37,000 were achieved for ^{137}Cs and ^{60}Co respectively. The concentrations of other radionuclides — ^{144}Ce , ^{125}Sb , and $^{95}\text{Zr-Nb}$ — were reduced to the analytical limits of detection (Table 1). The DF's for ^{106}Ru and ^{60}Co were 10 to 1000 times higher than those obtained previously in the ORNL waste development program.¹² The composition of the product water met U. S. Public Health

*Nuchar C-190, a product of West Virginia Pulp and Paper Co., Chemical Division, New York, N. Y.

Table 1. Product-Water Analysis of Low-Salt-Content Waste Treated by the Water Recycle Process^a

A. Overall Decontamination Factors (DF's) for Radionuclides

	Individual Isotope								Total Rare Earths	Gross Gamma	Gross Beta	Bulk ions (specific conductance)
	¹⁴⁴ Ce	⁶⁰ Co	¹³⁷ Cs	¹³¹ I	¹⁰⁶ Ru	¹²⁵ Sb	⁹⁰ Sr	⁹⁵ Zr-Nb				
Waste analysis, dis min ⁻¹ ml ⁻¹ or counts min ⁻¹ ml ⁻¹	28.0	245	162	277	96	3.4	55.1	1.7	52.0	254	198	80 ^b
DF's for product water	800 ^c	37,000	14,000	3,600	1230	210 ^c	5700	350 ^c	1900	270 ^c	260 ^c	200 ^b

B. Wet Analysis

	Turbidity (JTU) ^d	Total Dissolved Solids (ppm)	Total Hardness (ppm CaCO ₃)	Total Alkalinity (ppm CaCO ₃)	Calcium Hardness (ppm CaCO ₃)	Na (ppm)	Al (ppm)	Si (ppm)	PO ₄ (ppm)	SO ₄ (ppm)	F (ppm)	Cl (ppm)	NO ₃ (ppm)	C (ppm)
Waste water	10	•	24	19.6	21	3	<0.1	0.56	0.17	<2	0.26	<1	3	38
Product water	<0.05	<1	0.1	<1	<0.1	<0.1	<0.1	<0.1	0.02	<2	<0.02	<1	<1	<2
USPHS standards ^f	5	500	•	•	•	•	•	•	0.1 ^h	250	1	250	45	1

^a Conditions: alum-silica-nonionic polyelectrolyte clarification; cation-anion exchange; sorption on vegetable-based activated carbon as a final polishing step. DF values are for product water with a specific conductance of less than 1 micromho/cm.

^b The dimensions of specific conductance are micromhos/cm.

^c These are minimum values, since all of the radioactivity in the waste water was reduced to the analytical limits of detection (background).

^d Jackson turbidity unit. Turbidities measured on low-angle turbidity meter. Ref: A. P. Black and S. A. Hannah, J. Am. Water Works Assoc. 57, 901 (1965).

^e Not determined.

^f U. S. Public Health Service Drinking Water Standards, 1962.

^g No limits given.

^h No limit given. This figure is considered to represent the safe tolerance level in drinking water for man. Ref: T. R. Camp, Water and Its Impurities, p. 93, Reinhold, New York, 1963.

Service specifications regarding permissible concentrations of radioactive elements¹³ (i.e., < 2% of the MPC for a 168-hr week if ^{131}I is excluded; < 10% of the MPC if ^{131}I is included) as well as nonradioactive elements.¹⁴ The salt content of the product water was reduced to less than 2 ppm, i.e., greater than a 20-fold reduction.

In the demineralization-carbon system, specific conductance was a simple indicator of resin-bed exhaustion, since the breakthrough of bulk ions has been found to precede breakthrough of the radionuclides (Fig. 6). Ion exchange treatment alone removed most of the ^{137}Cs and ^{90}Sr , but sorption on activated carbon was required in order to obtain high DF's for cobalt and ruthenium. The DF for demineralization, as indicated by specific conductance measurements (Table 1), was 200.

The regeneration of the cation exchange column required six resin volumes of 4 M HNO_3 to reduce the activity level of the radionuclides on the resin to 0.1% of the level of the sorption cycle; the first three bed volumes (BV's) containing the bulk of the activity were diverted to intermediate-level waste, while the last three BV's were retained for a subsequent regeneration. In similar fashion, 8 BV's of 2 M NaOH were required to reduce the concentrations of the radionuclides on the anion exchange column to 1% of the original. Cobalt-60 was removed from the activated-carbon column by using 0.5 M HNO_3 as the eluent; 4 BV's were required to reduce the activity resulting from this radionuclide to 0.1% of that originally present.

Important parameters that could significantly affect the process operation in recycle water treatment include the pH of the raw waste, the specific conductance of this waste, the synthetic-detergent content of the waste, and the effect of increased amounts of contaminants that may accumulate during recycle. The pH of the raw waste must be in the range of 7 to 8 prior to coagulation-clarification treatment. This allows proper alum floc formation, which is essential to optimum removal of the radiocolloids, especially ^{106}Pu and ^{60}Co . The required alum dose is more dependent on the specific conductance of the waste than on the turbidity of the waste; ORNL-LLW with a specific conductance of 250 to 300 micromhos/cm and about 15 Jackson

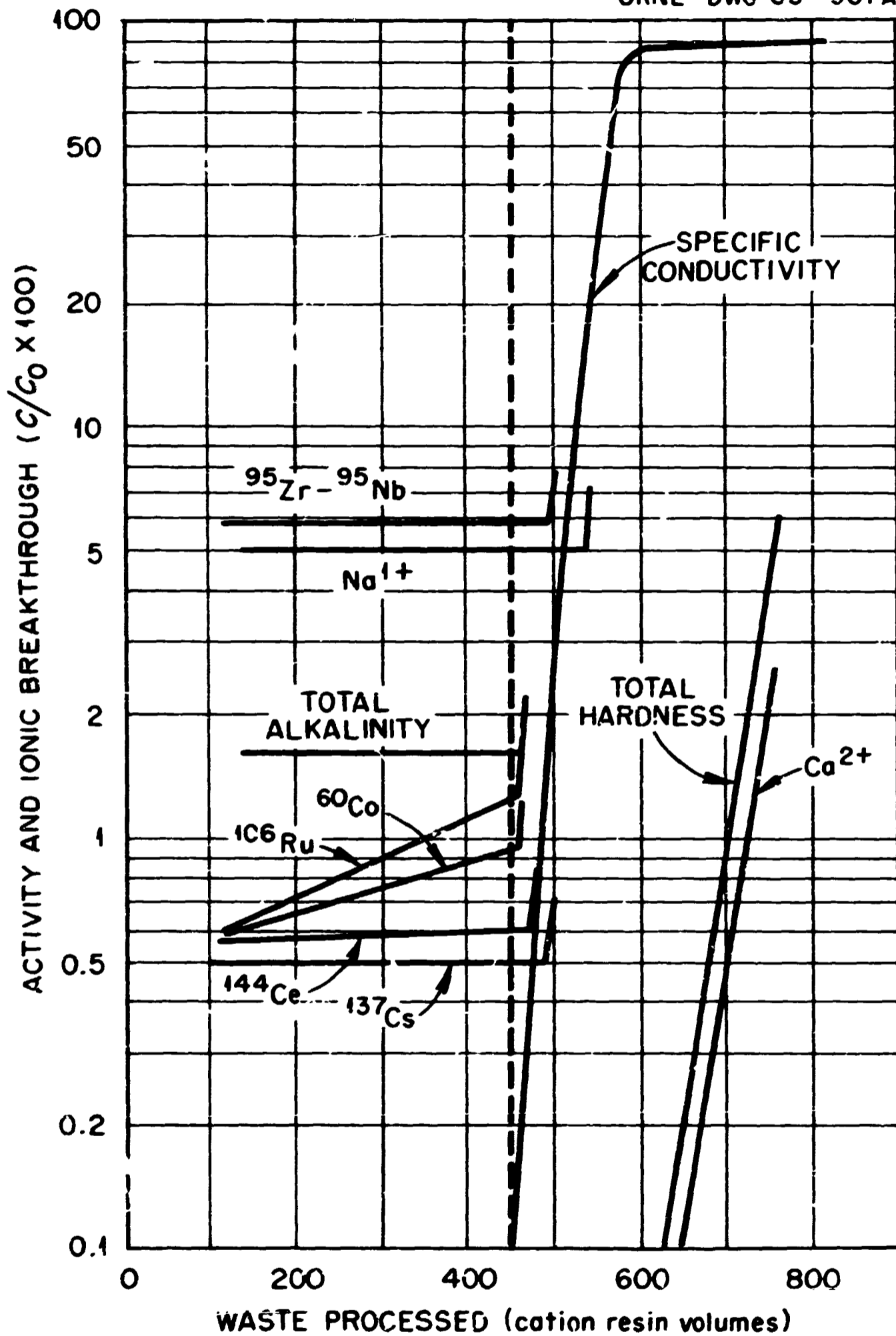


Fig. 6. Breakthrough of Bulk Ions and Radionuclides. Specific-conductance measurements signal imminent breakthrough of radionuclides because bulk ions break through first.

turbidity units (JTU) required 20 ppm $\text{Al}_2(\text{SO}_4)_3$, while recycle waste with a specific conductance of 50 to 60 micromhos/cm and turbidities ranging from 5 to 25 JTU required only 2 to 3 ppm $\text{Al}_2(\text{SO}_4)_3$ (i.e., about one-tenth of the dose required for ORNL-LLW). When large concentrations of synthetic detergent (syndets) are present in waste, the addition of a medium-long-chain cationic polyelectrolyte may be necessary in order to optimize clarification. This polyelectrolyte serves to decrease the negative ZP of the suspended particles sufficiently to permit alum and activated silica to neutralize the surface charges. In one micro-pilot plant run, 10-ppm quantities of tripolyphosphate (TP) and alkylbenzenesulfonate (ABS) were added to the recycle waste; the combined concentration of these compounds is comparable to the maximum syndet concentration in secondary effluent water, as reported by the U.S. Public Health Service.¹⁵ In the coagulation step, approximately 20 ppm of Primafloc C-3,* approximately 15 ppm of $\text{Al}_2(\text{SO}_4)_3$ and 0.2 ppm of activated silica (as SiO_2) were added. The addition of larger quantities of coagulants and coagulant aids significantly increased the total dissolved solids present in the waste and resulted in the reduction of the ion exchange capacity for treating the waste by 33% (i.e., to about 1800 BV's).

3. LABORATORY DEVELOPMENT OF THE PROCESS

Much of the laboratory work connected with the development of the water recycle process was devoted to a careful examination of the conditions necessary to achieve optimum clarification of raw wastes on a continuous basis, since the removal of the ionic radioactive contaminants could be readily accomplished via cation-anion exchange methods.

One of the most significant and controlling factors in the clarification of waste waters is ZP control.¹⁶ Although several other factors must also be taken

*A medium-long-chain polyamine of the cationic type, with a molecular weight of 10,000 to 20,000; product of Rohm and Haas Co., Philadelphia, Pa.

into account, it is essential to neutralize the mutual repelling forces (ZP) of the colloids since they keep the particles in suspension and hinder or prevent agglomeration. Coagulation is achieved when a sufficient quantity of alum and other coagulant aids is added to encapsulate all the colloidal particles in hydrated aluminum oxide. Zero ZP (with a tolerance of ± 5 mv) is attained when the amphoteric aluminum oxide floc is at a pH near the isoelectric point.

Exploratory jar (or beaker) tests were made with ORNL-LLW to identify the optimum conditions needed to clarify waste water, using ZP control. Each test consisted of the following steps: (1) 2 min of flash mixing (1750 rpm) of the waste water after alum was added; (2) 15 min of flocculation at 60 rpm, followed by 15 min of mixing at 20 rpm; and (3) 30 min of settling time. Electrophoretic mobility measurements were made, immediately after the flash-mixing step, with the use of a Zeta-Meter;* these measurements were subsequently converted to ZP values.

The average ZP (using the above method) of samples of ORNL-LLW taken over a six-month period varied from -12 to -18 mv (Fig. 7); this range is slightly less negative than that for suspensions in raw water (reported¹⁷ to be -16 to -22 mv). Test objectives included the determination of: (1) the amount of alum needed to bring the ZP of the waste to zero, (2) the influence of other coagulant aids on zeta potential when they were added along with alum, and (3) the effect of speed on flash mixing. Figure 8 shows that 10 to 20 ppm of alum is needed to neutralize the negative ZP for colloids (in waste) that have initial ZP values ranging from -11 to -18 mv.

The effects of coagulant aids on ZP (Table 2) are as follows:

1. Grundite clay, which can be added to give bulk to the light alum floc and to decrease subsequent settling time for clarification, tends to decrease the amount of alum subsequently needed to attain zero ZP. (The ZP of Grundite clay in tap water is about -10.8 mv, which is less negative than the average ZP of the suspensions in ORNL-LLW.)

*Product of Zeta-Meter, Inc., New York, N. Y.

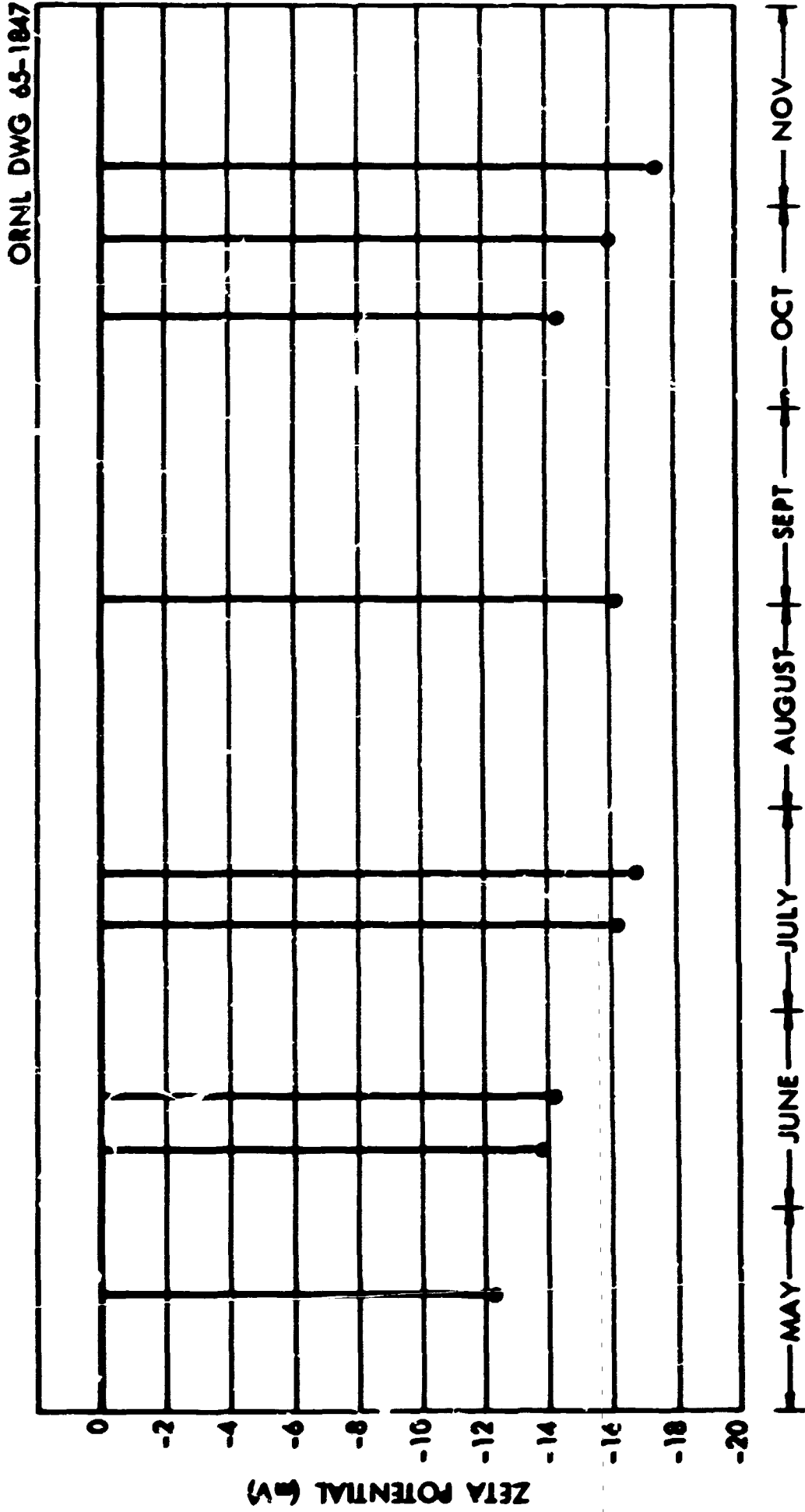


Fig. 7. Zeta-Potential Values for Samples of Kaw, Low-Level Radioactive Waste Taken in 1964.

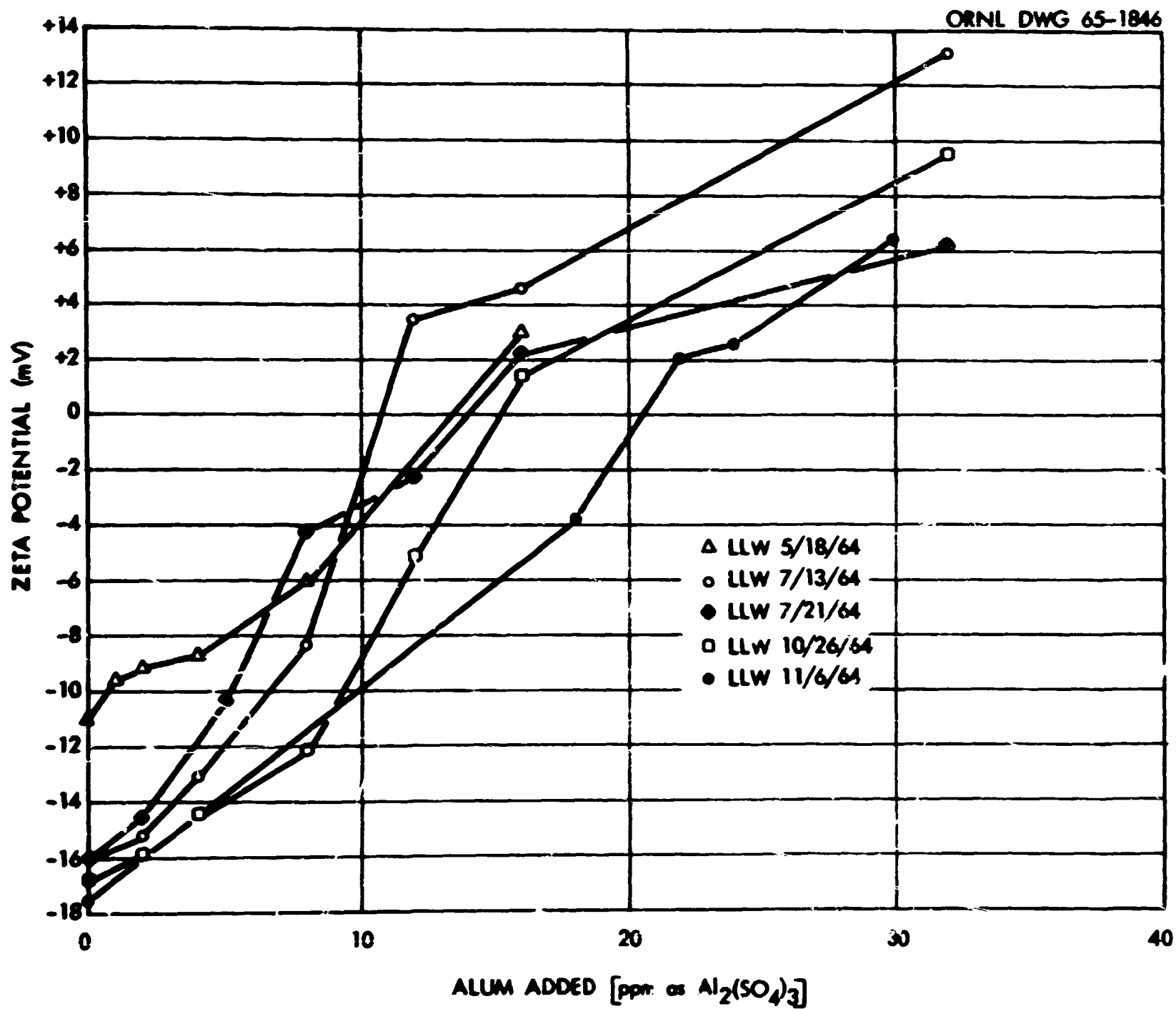


Fig. 8. Alum Coagulation of LLW, Using Zeta-Potential Control.

Table 2. Changes in Zeta Potential When Coagulant Aids Are Used in Conjunction with Alum Clarification

Date ORNL-LLV Sample Was Taken	Coagulant Aid		Alum Conc. (ppm)	Zeta Potential (mv)	
	Type	Purpose			
7-13-64	Grundite clay	To densify alum floc	0	0	-16.2
			0	13	0
			10	13	0
			20	12	0
			40	11	0
			80	10	0
11-6-64	Activated silica	To toughen fragile alum floc	0	0	-14.8
			0	24	+5.2
			0.5	24	+2.2
			1.0	24	-2.3
			2.0	24	-5.8
			5.0	24	-10.2
			10.0	24	-12.5
			15.0	24	-14.2
7-13-64	Primafluc C-3 ^a	To aid in agglomeration during flocculation	0	7	-5.7
			0	7 ^b	-8.0
			0.5	7 ^b	+3.1
			1.0	7 ^b	+8.2
			1.5	7 ^b	+11.2
			2.0	7 ^b	+12.7

^aA cationic, medium-length-chain polyamine with a molecular weight of 10,000 to 20,000; product of Rohm and Haas Co., Philadelphia, Pa.

^b7 ppm of alum and 40 ppm of Grundite clay.

2. Activated silica, which can be added to toughen the fragile alum floc, increases the amount of alum subsequently needed to attain zero ZP. (The ZP of activated silica in tap water is about -15 mv, which is about the same as that of the suspensoids in ORNL-LLW.)
3. Additions of small amounts of organic polyelectrolytes such as Primafloc C-3, which can be added to the waste water after the flash-mixing step to assist in floc agglomeration, produce greater changes in ZP than inorganic coagulants such as alum; for example, only 0.5 ppm of Primafloc C-3 shifted the ZP of the suspended particles in LLW from about -6 to +3 mv (Table 2).

Microscopic examination of the aluminum hydroxide floc showed that flash mixing at 1750 rpm caused the alum to be distributed more rapidly and more uniformly, and resulted in a more-complete enmeshing of the suspensoids, than mixing at 200 to 300 rpm (assuming that 300 rpm is the maximum speed obtainable with a standard laboratory stirrer). This result agrees with the work of Riddick,¹³ who reported that, in the first one-thousandth of a second following the addition of alum, the hydrous oxide polymer either encapsulates a colloid or unites with another alum polymer. The latter action results in useless floc formation.

Optimum clarification conditions were established by determining the amount of solid that remains suspended in the supernate in the jar tests. After a settling time of 30 min, 250 ml of the supernate in each jar was filtered through a 0.45- μ millipore filter, and the particulate concentration on the filter was determined. Best results were obtained when $Al_2(SO_4)_3$ and SiO_2 were added successively to give respective concentrations of 20-30 and 0.5 ppm (Fig. 9). In this case, the average ZP of the suspended solids in the waste increased from an original -14 to -17 mv to +2.5 mv. After this treatment, only 20 particles were found to be present per square centimeter of membrane filter area per milliliter of clarified waste, as compared with 300 particles for the raw waste.

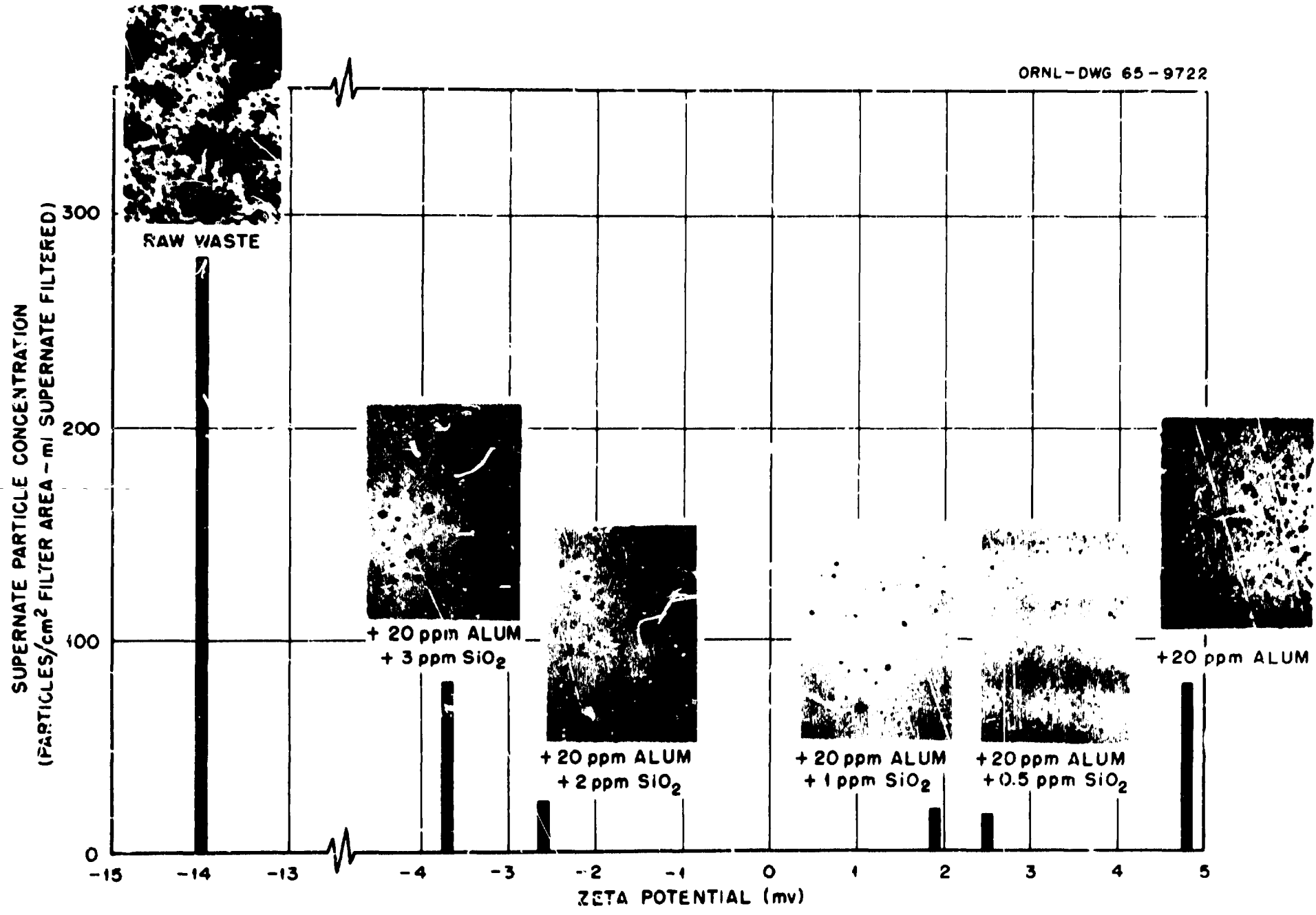


Fig. 9. Solids Filtered out of Treated Waste Supernatant onto a Membrane Filter at a Zeta Potential of +2.5 mv. Jar-test conditions: 30 min of flocculation followed by 30 min of settling.

The clarification of recycle waste water containing a low concentration of total dissolved solids (i.e., a specific conductance of ~ 50 micromhos/cm) had to be treated differently from LLW; this water required only about one-fifth to one-tenth of the quantity of alum used in the treatment of ORNL-LLW having a concentration of total dissolved solids similar to that of tap water (i.e., a specific conductance of ~ 300 micromhos/cm). Both activated silica and a nonionic polyelectrolyte were needed as coagulant aids in order to improve the settling characteristics of the alum floc formed. Neither the addition of a polyelectrolyte nor an alum-activated silica treatment alone produced a clarified product of the quality obtained by using the activated silica and polyelectrolyte in combination as coagulant aids.

Jar tests at pH 7, which is the optimum pH for alum floc formation, demonstrated that, in the presence of a polyphosphate (a constituent of syndets), alum flocculation and clarification are difficult. This is particularly true for water having a low calcium content (e.g., recycle water). The formation of alum floc was studied in distilled water containing 25 to 30 ppm of ortho-, pyro-, tripoly-, or hexameta-phosphate (as PO_4^{3-}). Alum floc was formed in orthophosphate solutions both in the presence and in the absence of calcium, and orthophosphate was removed (Fig. 10). In polyphosphate solutions containing 60 ppm of $\text{Al}_2(\text{SO}_4)_3$, no floc was formed in the absence of calcium. With the addition of 30 ppm of Ca^{2+} , an alum floc was formed immediately, and 80 to 95% of each of the polyphosphates was removed. Results of detailed column sorption tests with activated alumina, which is dehydrated alum floc (with less surface area), indicate that polyphosphate sorption was significantly higher when the calcium/phosphate mole ratio was 1.0 or higher; jar tests were made under conditions where these ratios varied from 0 to 3.0. Thus, when polyphosphates are present in recycle water, the calcium/phosphate mole ratio should probably be at least 1.0 in order to achieve optimum clarification with a minimum amount of alum.

4. CONCLUSIONS

Micro-pilot plant tests of a water recycle process demonstrated the feasibility of using a closed-cycle method to treat low-level radioactive waste having a low salt content. This process would allow the return of demineralized, well-decontaminated recycle water to a nuclear installation for reuse. It represents an improvement over the traditional "treat and discharge" methods of treatment. The flocculation-clarification treatment reduced the solids content (turbidity) of ORNL-LLW from 10 to less than 0.05 JTU, a level that is comparable to that of high-quality distilled water. Overall decontamination factors of 1000 to 10,000 were obtained for all major radioactive species up to 2400 volumes of waste treated per bed volume of cation resin. The product water met U. S. Public Health Service standards for the radioactive elements determined (i.e., less than 2% of the MPC for a 168-hr week if ^{131}I is excluded; < 10% of the MPC if ^{131}I is included). Specific conductance can be used as a simple indicator of resin-bed exhaustion since the breakthrough of bulk ions has been found to precede breakthrough of radioelements. Before breakthrough, the DF for demineralization was 200.

5. ACKNOWLEDGMENT

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