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**A Research Report for the
Atlantic Richfield Hanford Company**

**Evaluation of a Precipitation-
Ion Exchange Process for
Treatment of Laundry Waste**

**B. W. Mercer
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March 15 1977

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EVALUATION OF A PRECIPITATION-ION EXCHANGE
PROCESS FOR TREATMENT OF LAUNDRY WASTE

by

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WATER AND LAND RESOURCES DEPARTMENT
WATER AND WASTE MANAGEMENT SECTION

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EVALUATION OF A PRECIPITATION-ION EXCHANGE PROCESS FOR TREATMENT OF LAUNDRY WASTE

B. W. Mercer and L. L. Ames

INTRODUCTION

A study was initiated by Battelle, Pacific Northwest Laboratories (PNL) at the request of the Atlantic Richfield Hanford Company to characterize the 200-West laundry wastewater and evaluate alternative treatment systems for this waste stream. The laundry wastewater is presently discharged without treatment to the U Pond via the U-14 Ditch. Although the radioactivity in this waste is less than recommended release guidelines,⁽¹⁾ a buildup of radioactivity occurs in the ditch and pond sediments because of the sorptive characteristics of the sediments. The 1976 analyses of 216-U-14 ditch solids, removed from the ditch and piled along the west banks, showed surface readings of up to 10,000 cts/min beta-gamma.^(a)

Treatment of the laundry wastewater is one of several approaches to controlling the discharge of radionuclides from this source. Other possible approaches include methods to control the radioactivity levels of soiled apparel laundered at the facility and alternate cleaning methods such as dry cleaning. Clearly, an attempt should be made to limit the discharge of radionuclides in the laundry wastewater to a level that can be economically achieved through the best available technology.

An added problem concerning the laundry wastewater discharges is the presence of plant growth nutrients such as phosphorus and nitrogen. Cleaning compounds used by the laundry contain substantial quantities of phosphates and ammonia. The discharge of these substances to U Pond contribute to the fertilization of plant growth in the pond, which, in turn, attracts wildlife.

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^a Memo from J. V. Panesko to distribution, Laundry Effluent Data, February 16, 1977.

The first part of this study concerns chemical and radiological characterization of the laundry wastewater and evaluation of alternative treatment processes to render the waste more suitable for discharge to surface receiving waters. The second part of the study involves bench-scale pilot plant testing of a precipitation-ion exchange process for treating the laundry wastewater.

SUMMARY AND CONCLUSIONS

Bench-scale pilot plant studies have been conducted to evaluate chemical coagulation and ion exchange for decontamination of 2724-W laundry wastewater. Chemical coagulation serves to remove suspended matter and scavenge radionuclides such as rare earth fission products and transuranic isotopes. Chemical coagulation is accomplished at high pH (pH 11) to avoid complexant problems and assure good transuranic radionuclide removals. A cesium and strontium selective zeolite, clinoptilolite, is used to remove cesium which is not precipitated during the coagulation step and to remove strontium which is only partially removed by coagulation.

A flowsheet was developed whereby the laundry wastewater is treated with Fe^{+3} , Mg^{+2} , and Ca^{+2} salts at pH 11 to coagulate suspended solids and scavenge radionuclides. Next, the coagulated wastewater is flocculated with the aid of a polymeric flocculant, and transferred to a sedimentation tank where most of the floc settles out and is drawn off as sludge. The clarified water is filtered to remove residual suspended matter and is then treated by ion exchange to remove cesium and residual strontium. Finally, the sludge from the clarifier is dewatered by gravity thickening followed by vacuum filtration to produce a damp cake for solid waste disposal.

The results of the pilot plant studies are summarized as follows:

- Decontamination factors of 70, for strontium, and more than 100, for cesium, were achieved under optimum conditions by chemical coagulation and ion exchange. Decontamination factors exceeding 90 were measured for europium, a substitute for rare earth and transuranic radionuclides, by coagulation with a combination of ferric chloride, magnesium chloride, and calcium chloride added to the wastewater at pH 11.
- Coagulation with ferric chloride, magnesium chloride, and calcium chloride to the wastewater at pH 11 was more effective for turbidity removal than coagulation with lime.
- Addition of up to 1.7 lb of clinoptilolite fines per 1000 gallons of wastewater during the coagulation step did not substantially increase strontium and cesium removal.

- Filtration of the laundry wastewater without chemical coagulation reduced suspended solids by only 25%. About 70% of the suspended solids remaining in the filtered wastewater were removed in the zeolite column causing plugging which could not be easily dislodged by backwashing.
- Plugging of the ion exchange columns by previously clarified wastewater occurred during the pilot plant studies and required short periods of limited backwashing to relieve the plug. Calcium carbonate was identified in one column and is believed to have resulted from post-precipitation during the several-day storage period for the clarified waste. Post-precipitation of calcium carbonate is not expected to be a severe problem in a full-scale plant where the detention time between filtration and ion exchange is brief. A high pressure surface wash should be included in plant size ion exchange columns to break up crust or plugs at the surface of the zeolite.
- Addition of sodium hypochlorite to the ion exchange column feed to control biological growths may cause nitrogen gas formation in the ion exchange columns.
- Sludges removed from the clarifier generally settled to 4 to 5% of the total wastewater volume within 3 hr. The sludge can be further dewatered by gravity thickening.
- Centrifugation of iron sludges for 2 min at 2000 g reduced the sludge volume to about 1% of the total wastewater volume. Centrifuged iron contained about 4% dry solids.
- Wet iron sludges from the sludge storage tank were readily dewatered by vacuum filtration in the laboratory. The filter cakes contained 9 to 11% dry solids. Vacuum filtration of this type of sludge appears to be superior to centrifugation.

WASTEWATER CHARACTERIZATION AND ALTERNATIVE TREATMENT PROCESS EVALUATION

In the characterization studies, samples of laundry wastewater were collected and analyzed for radioactive and nonradioactive constituents.

RADIOACTIVE CONSTITUENTS OF THE WASTEWATER

Eight samples of laundry wastewater were collected for the characterization studies during the months of April, May, and September 1976. The samples were taken from a common sump which receives wash and rinse water from the mask cleaning facility, the radioactive apparel laundry and the nonradioactive apparel laundry. The nonradioactive apparel laundry introduces approximately 20% dilution to the radionuclide content of the other two sources. The samples were collected on a random schedule over a period of several hours during the date of sampling to assure a composite of the wastewater discharged during that period. Analyses of samples of laundry wastewater collected for the pilot plant studies during the period November 29, 1976 through January 14, 1977 are also included in the characterization results. These samples were collected over a one-half hour period and thus may not be as representative as earlier samples.

The results of the radiochemical analysis are presented in Table 1. The amount of suspended solids in the samples varied from 54 to 330 mg/l, much of which appeared to be very finely divided or colloidal. Characterization samples taken during the period August 11, 1976 through August 26, 1976 were analyzed for ^{90}Sr and $^{239,240}\text{Pu}$. No detectable $^{239,240}\text{Pu}$ was found in this series of samples, while the ^{90}Sr varied from below the detection limit to 250 pCi/l.

NONRADIOACTIVE CONSTITUENTS

Physical-chemical or biological treatment is highly dependent on the physical-chemical characteristics of the laundry wastewater. Therefore, an extensive characterization of this waste stream was undertaken to establish the pertinent physical-chemical parameters. The results of laboratory analysis of the eight samples are given in Table 2.

TABLE 1. Radioactivity Results on Laundry Wastewater Samples

Characterization Wastewater Sample	Radioactivity, pCi/l								
	Total Alpha	239,240 _{Pu}	Total Beta*	90 _{Sr}	137 _{Cs}	141 _{Ce}	103 _{Ru}	59 _{Fe}	60 _{Co}
4/23/76 Solution	<40	-	1300	-	570	-	-	-	-
4/23/76 Solids	-	-	-	-	110	68	130	230	540
4/30/76 Solution	<40	-	290	-	100	-	-	-	-
4/30/76 Solids	-	-	-	-	23	230	-	-	23
5/5/76 Solution	180	-	620	-					
5/5/76 Solids	44	-	3700	-					
8/11/76 Solution	330	<28	870	20					
8/11/76 Solids	370	<13	1800	<20					
8/19/76 Solution	<14	-	430	230					
8/19/76 Solids	23	<11	560	210					
8/20/76 Solution	210	<10	680	250					
8/20/76 Solids	51	<5	890	100					
8/24/76 Solution	120	<10	510	200					
8/24/76 Solids	130	<11	1000	60					
8/26/76 Solution	16	-	110	20					
8/26/76 Solids	55	<6	770	<8					

Pilot Plant Feed Sample	Total Alpha	Total Beta
11/29/76 Total	<50	15,000
11/30/76 Solution	<60	2,000
11/30/76 Solids	<60	2,900
12/9/76 Solution	<60	600
12/9/76 Solids	<60	600
1/5/77 Total	<50	90
1/6/77 Solution	<60	200
1/6/77 Solids	<60	500

**As ¹⁰⁶Ru.

TABLE 2. Completed Physical-Chemical Analyses of Laundry Wastewater Samples
Concentrations in mg/l except pH and turbidity.

Date	BOD	COD	NH ₃ -N	Total P	Total	CO ₃ -	HCO ₃ -	Suspended Solids	Na ⁺	K ⁺	Mg ⁺²	Ca ⁺²	NO ₃ -N	Cl ⁻	SO ₄ -	Turbidity JTU	pH
4/23	84	198	45	44	702	532	170	91	310	2.4	4.7	18	0.11	4.3	17	260	9.9
4/30	270	626	12	48	780	660	120	330	350	3.1	2.5	9.0	0.25	2.0	16	>1000	10.1
5/5	120	177	27	16	465	308	157	79	190	1.6	2.8	8.9	0.04	<1.0	7.0	130	9.7
8/11	36	75	27	46	333	176	157	58	140	1.6	3.2	14	--	--	--	67	9.3
8/19	36	88	35	30	354	280	74	72	150	2.2	2.4	11	--	--	--	130	9.9
8/20	27	70	24	35	324	220	104	60	150	2.1	2.5	11	--	--	--	70	9.9
8/24	76	99	29	53	336	200	136	56	110	2.1	3.0	12	--	--	--	130	9.7
8/26	78	98	32	20	338	224	114	54	130	1.7	2.6	11	--	--	--	110	9.9
11/30	--	--	--	53	--	--	--	72	200	--	7.6	13	--	--	--	44	--
12/9	--	--	--	65	--	--	--	66	110	--	3.4	12	--	--	--	26	--
1/6*	--	--	--	107	--	--	--	111	307	--	4.2	20	--	--	--	55	--

*Year 1977. All other dates are in 1976.

Biochemical oxygen demand (BOD) and chemical oxygen demand (COD) indicate the concentration of oxidizable material (e.g., organics) present in the wastewater. Under the proper conditions material exerting a BOD can be removed by a biological process which converts the material to a sludge and CO_2 . The presence of much BOD in a wastewater can be a problem in a physical-chemical process because of the formation of slimes and other biological growths which feed on the BOD and adversely affect the hydraulics of the system. The BOD and COD values in the wastewater are in most instances less than those found in typical domestic sewage.

The ammonia nitrogen levels found in the waste are on the average slightly higher than the levels in domestic sewage, but the phosphorus levels are higher by a factor of 3 to 4. The waste, therefore, has a good fertilizer value for plant growth. The nitrate-nitrogen concentrations were determined in the samples taken in April and May and found to range from 0.4 to 0.26 mg/l. Because of these low levels, additional nitrate determinations were not performed. Sulfate and chloride analyses were also eliminated from the remaining characterization work since these parameters were not expected to change significantly and are not important to process evaluations. Phosphorus concentrations in the waste are significant not only for nutrient value but also for reactions with coagulating chemicals such as alum, iron salts, and lime.

Alkalinity is an important parameter with regard to chemical coagulation of the wastewater. The total alkalinity was found to vary by a factor of over 2 for the samples analyzed. The addition of alum or iron salts introduces acidity which is neutralized by the alkalinity present in the waste. If a high pH is desired, for example, as in coagulation with iron salts, it may be necessary to add alkali to the waste. Conversely, acid addition may be necessary with alum coagulation to attain the optimum pH for floc formation. Lime addition will result in the precipitation of calcium carbonate, the quantity of which will be a function of the carbonate-bicarbonate alkalinity. The precipitations of calcium carbonate and calcium hydroxyapatite (calcium phosphate) are important strontium removal mechanisms. However, underdosing or overdosing with lime can adversely affect

strontium removal. The optimum dosage of lime for strontium removal is a function of both the alkalinity (including carbonate-bicarbonate) and the phosphate concentration. Alkalinity also imposes restrictions on the use of membrane processes, reverse osmosis and electrodialysis. Acid addition is frequently necessary to adjust the pH to a range of 5 to 7 for optimum performance of these processes.

The concentrations of sodium, potassium, magnesium, and calcium in the waste are important for several reasons. Considering ion exchange as a treatment method must include the effect of these ions on the removal of radionuclides. The concentration of calcium and magnesium, for instance, will largely control the breakthrough capacity of a cation exchange resin for strontium. Calcium and magnesium also enter into chemical coagulation processes since the addition of alkali will cause both to precipitate. As previously stated, the calcium precipitates as the carbonate and/or apatite while the magnesium precipitates as the hydroxide. Magnesium hydroxide precipitation is often of considerable benefit in chemical coagulation of wastewater. The total cation concentration will also control the breakthrough of cesium and strontium through a cation exchange resin bed. In the case of cesium-selective ion exchangers, the potassium and ammonium ions are more significant than the other cations since these ions compete more strongly with the cesium ion.

Suspended solids (SS) concentrations in the wastewater were found to vary from 54 to 330 mg/l. The high SS value coincided with a high turbidity value (>1000 JTU) and may have resulted from laundering excessively soiled protective apparel and/or the use of large amounts of soda ash-clay mixture in the laundering process. Much of the suspended solids is finely dispersed and appears to be difficult to remove by filtration without the benefit of chemical coagulation.

PROCESS EVALUATION

Processing schemes which may be used for treating the laundry wastewater may employ one or several of the following:

- Ion Exchange
- Sedimentation

- Coagulation
- Filtration
- Evaporation
- Reverse Osmosis
- Electrodialysis
- Adsorption
- Biological Treatment
- Precipitation

A discussion of each of these processes is presented below.

Ion Exchange

Ion exchange is a very effective method for removing radionuclides from certain types of wastewaters. In addition to achieving high decontamination factors, ion exchange is also capable of concentrating the radionuclides into very small volumes to minimize the cost of disposal. Many specialized ion exchangers have been developed which have high specificity for particular radionuclides in the presence of high electrolyte concentrations.

Despite the many advantages of ion exchange for treating radioactive wastewater several limitations must be considered prior to any attempt to use this process. These limitations include:

1. Radionuclides not in an ionic or charged form cannot be removed. These nonremovable forms include certain radiocolloids, radioactive particulate matter and soluble non-electrolyte forms. If not mechanically removed by filtration these non-exchangeable radionuclides will leak through the ion exchange bed reducing the decontamination efficiency of the process.
2. The concentration of suspended solids in the wastewater must be very low or plugging will render the ion exchange column unusable in many cases. It is generally not a good practice to use an ion exchange column both as a filter and ion exchanger. Although much particulate matter may be removed by backflushing, agglomeration or caking of the resin beads frequently occurs which destroys the effectiveness of the resin. The resin beads are also susceptible to blinding from oil and grease which are present in the laundry wastewater.

3. The concentration of dissolved electrolytes must be relatively low to attain high loading of radionuclides on the exchange column. About 2500 mg/l is generally accepted as the maximum total solids concentration although less than 1000 mg/l leads to more efficient use of the resin.⁽³⁾
4. In the case of mixed fission products a low pH is generally required for good removal by cation exchange resins. For a highly alkaline waste solution this may require costly addition of acid to attain the optimum pH. An estimated average of about 400 lb of 60% HNO_3 would be required daily to acidify the 50,000 gallons of wastewater generated.

Since much of the radioactivity is associated with the suspended solids in the 2724-W laundry waste, direct use of ion exchange does not appear feasible. It is anticipated that column plugging and high leakage of radionuclides will be problems. The waste contains both finely suspended matter which could pass through an ion exchange column with its sorbed radioactivity and larger particles which can plug the column. Combined filtration-ion exchange resins (powdered resins) are used to remove both soluble and particulate impurities in nuclear reactor water systems.⁽⁵⁾ However, the water is very high purity and the resins have a relatively long operating life which permits discarding them after exhaustion. The use of these powdered resins for treating low purity wastewater would be quite costly. In addition, the powdered resin beds are very shallow (~1/8 in.) and do not achieve high decontamination factors.

The effective use of deep resin beds for treating the laundry wastewater will require pretreatment to reduce the suspended solids content. Chemical coagulation, sedimentation and filtration is a very effective sequence of processing steps to prepare a clear feed stream to the ion exchange column. These steps are also effective for removing radioactive particulate matter and scavenging much of the soluble radioactivity from solution. The main disadvantage of chemical coagulation is the sludge produced which must be dewatered and disposed of. Chemical coagulation will be discussed in further detail in a later section.

Alternative methods of removing suspended matter from wastewater prior to ion exchange treatment include direct filtration with porous material such as felt or cloth; wire mesh sintered materials; membranes; and diatomaceous earth. As will be discussed in the section on filtration, none of these filters are considered satisfactory for feed preparation to ion exchange columns.

A further consideration in the use of ion exchange concerns the disposal of spent regenerant solutions or possibly the ion exchange material itself. Based on equilibrium data for a macroreticular, sulfonated, polystyrene-divinyl benzene cation exchange resin⁽⁶⁾ a daily flow from the laundry could be expected to be processed with a 100 to 250 gallon bed of this resin to breakthrough of cesium or strontium. Regeneration and rinsing of the ion exchange bed would produce an estimated 1000 to 2500 gallons of regenerant waste and rinse water for disposal.

Selective ion exchangers may be used in place of conventional organic resins to increase the volume out through the exchange beds to breakthrough. As indicated earlier, removal of a variety of radionuclides from wastewater will probably require acidification to use a selective ion exchanger such as sodium titanate. There are, however, several ion exchangers which can selectively remove cesium from neutral or basic solutions. These include the natural zeolites, clinoptilolite, chabazite, erionite, and mordenite. Mordenite is produced commercially as a synthetic zeolite which has been successfully used in the 200 Areas for removing cesium from condensate wastes. It is a relatively expensive zeolite, costing approximately \$2 per pound. Ion exchange grade clinoptilolite costs considerably less at about 15¢ per pound. Both of these zeolites are excellent cesium-selective ion exchangers. Clinoptilolite also performs well for removing strontium, providing competitive ions such as NH_4^+ , K^+ , and Ca^{+2} are not present in substantial concentrations.

In summary, ion exchange could be effective for treating laundry waste. However, the problem of clarifying the feed to the ion exchange bed complicates this approach. Direct filtration without chemical coagulation is normally a very difficult task for wastewater with relatively high and

variable suspended solids concentrations. In addition, low or moderately selective ion exchange resins (e.g., sulfonated polystyrene types) are too expensive for one-time use with laundry waste and for repeated use would require relatively large volumes of regenerant and rinse water which must be disposed of. A low cost cesium selective exchanger may be successfully used in conjunction with another process (e.g., chemical precipitation) which removes most of the radioactivity except radioactive cesium isotopes.

Sedimentation

Sedimentation without chemical pretreatment is an effective treatment step for those wastewaters which contain sufficient settleable solids to warrant their removal. Samples of 200-West laundry wastewater have had only minor quantities of settleable solids other than sand and gravel. Removal of the sand and gravel is normally accomplished in a grit chamber rather than a settling basin. Effective removal of suspended solids in the laundry waste will require chemical coagulation of the fine particulate matter contained in this stream. Coagulation serves to collect the smaller particles into large ones which will settle at a relatively rapid rate. Proper selection of the chemical coagulants can also aid in removing additional radionuclides from the waste solution by scavenging or coprecipitation of radionuclides. The subject of chemical precipitation, coagulation and filtration will be discussed in the next section.

A variety of clarifiers are available for sedimentation of suspended solids. These range from conventional circular clarifiers which have been used for many years in municipal and industrial waste treatment plants to modern high rate tube settling devices. Selection of a clarifier for a particular treatment scheme is often determined by pilot plant testing if previous experience is unavailable for the type of wastewater under study.

Coagulation

The removal of suspended matter is generally accomplished by one or more of three processes: 1) sedimentation, 2) flotation, or 3) filtration. Sedimentation and filtration are more commonly used for this purpose than flotation. To effectively use sedimentation and filtration, the particulate

matter must be sufficiently large to settle in a reasonable time period or become entrapped in a filter media. Since a substantial portion of the suspended matter in the laundry wastewater is too fine for rapid sedimentation or easy filtration, these particles must be agglomerated by chemical means. Aluminum and iron salts and lime are common coagulants for this purpose. In addition to aiding the removal of radioactive particulate matter, these coagulants form precipitates which are effective scavengers for many radionuclides.

Removal of particulate matter in wastewater is typically accomplished by adding a coagulant chemical such as ferric chloride to the wastewater in a rapid mix tank. Caustic or acid may also be added at this point to adjust the pH to the desired level. A coagulant aid such as long-chained organic polymers are also frequently added to aid in flocculating the precipitated and coagulated solids. The mixture is then routed to a flocculating tank where it is gently agitated or stirred to allow agglomeration of the solids into large flocs. Following the flocculation step, the mixture may then be routed to either a sedimentation basin or directly to a filter. Generally, mixtures with large quantities of floc will be settled prior to filtration since rapid plugging of the filter would occur in this case. Sedimentation basins are normally equipped with sludge removal devices that direct the settled sludge to a drain where it is pumped to a collection tank for further treatment.

Chemical coagulation or scavenging precipitation have been successfully used at many nuclear installations throughout the world for decontaminating radioactive wastewater. For example, according to M. Mass of Rockwell International, Rocky Flats currently uses ferric hydroxide precipitation at high pH to treat laundry and other wastewater produced at this installation. A rotary vacuum filter is used to dewater the sludge to about 60% moisture. Commercial automated units are available which are equipped with chemical feed systems, flocculating and sedimentation tanks, and a filter. These units are programmed to operate with a minimum of operator attention.

Filtration

Judging from the difficulties in the laboratory, direct filtration of the laundry waste is not recommended. Several 0.45 micron filters were used and considerable time was required to filter a liter of this wastewater. Probably the best removal with direct filtration would be obtained with diatomaceous earth filters. However, past experience with this type of filter for wastewater with variable suspended solids concentration has not been entirely satisfactory.⁽⁷⁻⁹⁾ In order to operate effectively, the body feed of diatomaceous earth must be varied with the solids content of the wastewater. To date there are no effective measuring devices which can sense the changes in suspended solids to allow proper adjustment of body feed. Turbidity can be monitored continuously. However this measurement is often not a good indicator of the suspended solids content.

Other filtering devices such as sand or multimedia filters and micro-screens are largely ineffective for removing small particulate matter. Membrane filters may be used effectively for removing fine particulate matter; however, operational costs for these units would be high.

Although direct filtration of the laundry wastewater might produce less sludge than a chemical coagulation process, further treatment by ion exchange, for example, would be required to remove a broad range of radionuclides. Thus the advantage gained by the small sludge volume would be lost by the increase in ion exchange regenerant volume.

Evaporation

Evaporation is widely used throughout the nuclear industry for decontaminating radioactive wastewater. With proper design, a very high decontamination factor (df) can be obtained for many radionuclides. The process is quite expensive, however, and would appear to be a case of overkill for 200-West laundry waste which would not require high dfs. In addition, foaming could be an adverse factor in the operation of an evaporator processing laundry wastewater.

Reverse Osmosis

Removal of both suspended solids and dissolved solids can be accomplished in a properly designed reverse osmosis (RO) unit. In order to avoid plugging the membranes with particulate matter, high turbulence is required over the surface of the membranes. The tubular RO unit is considered by many to be the best type for processing high solids wastewater. Reverse osmosis would probably be less costly than evaporation for desalination (and decontamination) of laundry wastewater. However, the dfs achieved by one stage of RO are much less than that of evaporation. Decontamination factors of 10 to 20 are common for RO, whereas evaporation can readily achieve a df of more than 10^3 in one stage.

Reverse osmosis produces two effluent streams: 1) a demineralized or decontaminated wastewater stream and 2) a concentrate stream containing most of the radioactivity and solids. The concentrate stream would require further treatment such as a second RO stage followed by chemical treatment and evaporation of the second stage concentrate. A capital cost of \$3,000,000 has been estimated for a facility using two-stage RO for the wash water, the first rinse water and the mask cleaning wastewater in addition to ion exchange for the remaining rinse water from the 200-West laundry.⁽¹⁰⁾ Much of the water in this processing scheme would be recycled for reuse.

The cost of RO for decontaminating 200-West laundry wastewater would be relatively high compared to the alternative of chemical coagulation, sedimentation, and filtration. Both processing schemes would require the disposal of solid waste if chemical treatment and evaporation is used for the RO concentrate. The quantity of dried solids produced by either processing scheme would range from 300 to 500 lb per day. The volume of dried solids cannot be accurately estimated at this time since it depends on the type of drier used for the wet sludge.

Electrodialysis

Electrodialysis (ED) is a membrane process which utilizes an electric potential to transport ions from a feed solution through ion exchange membranes to a concentrate stream. As with RO it produces a demineralized

water stream and a concentrate stream. Suspended solids are not removed by ED; therefore, any activity retained by these solids will not be removed. The decontamination efficiency of ED is limited with weak electrolyte solutions since insufficient electric current can be applied at reasonably low voltages to effect good electrolyte removals. The concentrate stream would also require the same treatment as the RO treatment scheme. Electrodialysis does not appear to be a viable candidate for laundry waste treatment.

Biological Treatment

Biological treatment is generally used to stabilize organic wastes; however, it has been found effective for removing radioactivity. Idaho Falls presently routes their laundry waste to the sanitary waste facility which employs activated sludge treatment. The wastes from this process are suitable for discharge to the environs, but the sludge is collected for disposal to a regulated burial ground. Biological treatment is generally not effective for removal of phosphorus and nitrogen.

Biological treatment can be accomplished by several different methods including activated sludge, trickling filters, and oxidation ponds. Activated sludge involves biological oxidation of organic matter, both soluble and suspended, through the use of a biological sludge which is contacted with the raw wastewater in aeration tanks. The sludge is separated from the wastewater in a clarifier sedimentation basin and a portion of the sludge is recycled to the aeration basin. The remaining sludge is either disposed of or treated further to stabilize it prior to disposal. Trickling filters accomplish the same objective, but the biological action takes place in a slime on the surface of a suitable media (e.g., rocks) placed in the basin. The raw wastewater is percolated over the media and oxygen is supplied by air passing through the media. The slimes which grow on the media eventually slough from the media and are collected as sludge in a clarifier. Oxidation ponds are frequently used where sufficient land is available to allow relatively slow biological oxidation to take place (i.e., oxygen is supplied by diffusion from the surface of the ponded water). Although biological treatment of laundry waste is feasible, the process has inherent disadvantages which do not favor its use in this situation. The activated sludge and

trickling filter processes cannot be shut down and restarted easily. A time period of several weeks is usually needed to establish optimum biological growth. In addition, only minimal phosphorus and nitrogen removal can be expected.

Adsorption

Adsorption of an activated carbon has been successfully used to remove radionuclides such as ^{60}Co and $^{95}\text{ZrNb}$ from low and intermediate level wastes.⁽¹¹⁾ The physical adsorptive properties of activated carbon would be expected to work well for removal of some organic complexed radionuclides and perhaps some radiocolloids. Activated carbon has not found widespread use for treating radioactive wastes and it is not expected to have much utility in the treatment of laundry wastewater for the removal of radioisotopes.

LABORATORY COAGULATION STUDIES

Jar tests were conducted to establish optimum chemical dosages to coagulate and flocculate suspended matter in the laundry wastewater.

CHEMICAL DOSE REQUIREMENTS

A series of 1-liter jar tests were carried out on two samples of laundry wastewater taken during the month of June 1976 when several Hanford production facilities were shutdown. Although the radionuclide content of these samples was low, it was believed that the chemical content was similar to that of normal operations. Therefore, chemical coagulation tests were expected to be the same as those for normal samples. The primary objective of this work was to determine the effects of various wastewater treatments on the removal of solids. In this work, the wastewater samples were used within 2 weeks of collection. A sample used approximately 5 weeks after collection did not give similar results for the same lime treatment. This behavior suggests that changes that are detrimental to common wastewater treatment methods take place in the wastewater if it stands for a long time at room temperature (22°C).

In most of the following jar tests, the treatment chemicals were added at 100 rpm of the stirrer. Common to all of the jar flocculation tests was the addition of 1 mg/l of an anionic flocculant aid.

After stirring at 100 rpm for 2 min, the stirrer was set at 30 rpm for 15 min and samples taken then for turbidity and other measurements. An exception was the lime coagulation tests where samples were taken at frequent intervals for turbidity measurement. Because the first laundry wastewater sample contained a red color, the lime jar tests also include color removal.

The jar test results with lime flocculation are listed in Table 3. Samples were taken at timed intervals for the various additions of lime. Not much change in turbidity occurred with time for the higher lime doses. The higher lime doses would be of interest here because of improved solids and phosphorus removals. Color was removed at 300 mg/l lime dose rate, but the pH climbed steadily with increasing lime content as well. Calcium and

TABLE 3. Jar Tests on Laundry Waste with Lime

Turbidity, JTU at -	Ca(OH) ₂ , mg/l						
	50	100	200	300	400	500	1000
	JTU						
1 min.	98	105	98	32	17	7.5	3.5
3 min.	95	108	102	25	15	7.5	3.5
5 min.	82	104	92	22	15	8.0	4.0
10 min.	73	91	82	21	14	11.0	6.0
20 min.	76	103	95	21	14	8.0	6.5
Color	Red	Red	Red	None	None	None	None
pH	10.70	11.05	11.45	11.70	11.80	11.85	12.20
Ca, mg/l	34	45	28	60	64	74	326
PO ₄ -P, mg/l	14	12	10	7	5	4	2.5
Mg, mg/l	2.6	1.8	0.7	0.22	0.13	0.14	0.07
Sr, mg/l	0.060	0.054	0.047	0.059	0.051	0.097	0.262
Influent Sr, mg/l	0.070	0.083	0.109	0.135	0.161	0.187	0.317
Influent Ca, mg/l	39	66	120	174	228	282	552
Influent PO ₄ -P - 19 mg/l							
Influent pH - 10.25							
Influent Mg - 2.6 mg/l							
Influent Turbidity - 150 JTU							
Temperature - 22°C							

TABLE 4. Jar Tests on Laundry Waste With FeCl₃.
In Each Case 40 mg/l Ca²⁺, 20 mg/l Mg²⁺,
and 100 mg/l Fe³⁺ Were Added in Common.

	NaOH Added, mg/l				
	0	60	120	200	400
JTU	0.3	0.2	0.2	0.2	0.3
pH	6.8	7.3	8.9	9.7	10.8
Ca, mg/l	52	--	--	14	4.6
Mg, mg/l	21.6	--	--	16	1.1
Sr, mg/l	0.071	--	--	0.015	0.012
PO ₄ -P, mg/l	12.5	10.5	10.0	13.0	17.0
Influent Ca - 52 mg/l					
Influent JTU - 150					
Influent pH - 10.25					
Influent Mg - 22.6 mg/l					
Influent Sr - 0.071 mg/l					
Influent PO ₄ -P - 18.5 mg/l					
Temperature - 22°C					

strontium removal were optimum at a lime dose rate of 200 mg/l. The influent calcium and strontium contents given in Table 4 include calcium and strontium contributed by the added lime. There was 0.057 mg/l Sr and 12.0 mg/l Ca in the original wastewater. About 80% strontium removal is the best that can be achieved with cold lime precipitation. Oak Ridge National Laboratory (ORNL) data with wastewater-lime precipitations confirm the 80% strontium removal is typical. As the lime dose rate approaches 1 g/l, product water hardness and pH rise considerably as the phosphorus, magnesium, and turbidity decline. The amount of magnesium, if any, in the added lime was not determined.

Some jar tests with FeCl_3 flocculant are also given in Table 4. The amount of iron added was constant but the amount of NaOH added was varied to affect pH. Calcium, magnesium, and strontium removal increased directly with solution pH, while phosphorus removal was nearly inversely related to pH. The influent values given for calcium and magnesium include the amounts added during chemical addition as well as those cations in the original wastewater. Similar tests with FeCl_3 flocculation are shown in Table 5. The best flocculation was with 60 mg/l FeCl_3 addition. Note that this is another waste with different turbidity and phosphorus content than the previous wastewater.

Jar tests with different dose rates of alum are given in Table 6. The turbidity is well removed even at the lowest dose rate, but considerable alum is required for good phosphorus removals. In addition, alum sludges are difficult to dewater as compared to lime sludges. However, lime produces a product water that is hard and high in pH. If ammonia removal by stripping is of little interest, iron and aluminum flocculants should be considered. Magnesium chloride was investigated as an aid to lime flocculations with the results shown in Table 7. Magnesium does not help much in phosphorus removal but does hold the product water pH down and can be used in turbidity removal.

Because the laundry waste ordinarily is thermally hot, the effects of temperature were examined. Temperature has little effect in the range investigated on pH or turbidity removal as seen in Table 8. However, phosphorus removal increases considerably. It is difficult to see why there is

TABLE 5. Jar Tests on Laundry Waste with FeCl_3 .

	<u>FeCl_3 Added, mg/l</u>		
	<u>20</u>	<u>60</u>	<u>100</u>
Turbidity, JTU	13	0.6	0.6
Floc Settling Rate	Medium	Fast	Slow
pH	8.8	6.8	5.2
$\text{PO}_4\text{-P}$, mg/l	14.0	7.0	4.5
Influent Turbidity - 34 JTU			
Influent pH - 9.55			
Influent $\text{PO}_4\text{-P}$ - 35 mg/l			
Temperature - 22°C			

TABLE 6. Jar Tests on Laundry Waste with Alum.

	<u>Alum Added, mg/l</u>		
	<u>80</u>	<u>160</u>	<u>240</u>
Turbidity, JTU	6.5	4	0.4
pH	8.9	8.4	7.5
$\text{PO}_4\text{-P}$, mg/l	25.0	16.0	10.5
Influent Turbidity - 34 JTU			
Influent pH - 9.55			
Influent $\text{PO}_4\text{-P}$ - 35 mg/l			
Temperature - 22°C			

TABLE 7. Jar Tests on Laundry Waste With MgCl_2 .
In Each Case, 200 mg/l $\text{Ca}(\text{OH})_2$ Was Added.

	Mg Added, mg/l		
	0	20	40
Turbidity, JTU	49	2.5	1.3
pH	11	10.9	10.8
$\text{PO}_4\text{-P}$, mg/l	16.0	14.0	11.0
Influent Turbidity - 34 JTU			
Influent pH - 9.55			
Influent $\text{PO}_4\text{-P}$ - 35 mg/l			
Temperature - 22°C			

TABLE 8. Jar Tests on Laundry Waste for Effect of Temperature. The Addition of 20 mg/l Mg and 200 mg/l $\text{Ca}(\text{OH})_2$ Was Common To Each Solution.

Starting Temperature, °C	22	35	50
Final Temperature, °C	22	33	43
Turbidity, JTU	1.7	2.1	1.3
pH	10.8	10.8	10.7
$\text{PO}_4\text{-P}$, mg/l	28.5	10.5	11.0
Influent Turbidity - 34 JTU			
Influent pH - 9.55			
Influent $\text{PO}_4\text{-P}$ - 35 mg/l			

an effect because everything else, except phosphorus content, remains the same. In addition, removal at 35°C is the same as at 50°C, which cannot represent a direct temperature effect.

Levels of radioactivity in the laundry wastewaters used in these experiments were too low to be of much value. Total alpha values in the influents were 0.24 ± 0.085 dpm/ml and 0.19 ± 0.076 dpm/ml, respectively. Total beta values for the influents were 0.44 ± 0.28 dpm/ml and 0.96 ± 0.33 dpm/ml, respectively. Gamma scans were not run because of the low activity. Product waters after the above treatments were less than 0.2 dpm/ml total alpha and less than 0.3 dpm/ml total beta. Laundry wastewaters containing more radioactivity than these samples will have to be used to obtain concrete results on process decontamination factors. The cold strontium results help to some extent to estimate process strontium radioisotope decontaminations that could be attained.

Alum and ferric chloride effected the best removals of suspended matter as evidenced by the low turbidities attained with these coagulations. Additional Ca^{+2} and Mg^{+2} were added to the experiments, the results of which are given in Table 4, to simulate the type of treatment used for low and intermediate level wastewater at Rocky Flats. The Ca^{+2} is reported to aid in plutonium removal while Mg^{+2} assists the Fe^{+3} in promoting good floc formation.

RADIOISOTOPE TRACER EXPERIMENTS

Five samples of laundry wastewater were collected during the month of August following return to normal operation. Three of the samples contained sufficient wastewater for characterization analysis and further jar tests for determining chemical coagulation efficiency. Trace ^{137}Cs , ^{85}Sr , and ^{144}Ce were added to portions of the samples used for the jar tests to measure the dfs obtained by the coagulation steps.

The character of the waste obtained in August was found to be somewhat different from that obtained in June. The dosage of alum required to clarify the August samples increased by a factor of 2. An alum dosage of 240 mg/l was sufficient to reduce the turbidity of the June samples to less than 1 JTU, whereas 480 mg/l of alum was required to reduce the turbidity of the

August samples to less than 1 JTU. Ferric chloride without Ca^{+2} , Mg^{+2} , and NaOH addition was similarly affected, whereas, treatment with lime was unaffected.

Preliminary results indicate that good clarification by coagulation with alum, lime, or FeCl_3 was very effective for ^{144}Ce removal (>90%). Alum coagulation was not adequate for either ^{85}Sr or ^{137}Cs removal. Ferric chloride with 40 mg/l Ca^{+2} and 20 mg/l added was good for both ^{144}Ce and ^{85}Sr removal (^{144}Ce >90% and ^{85}Sr ~85%). Lime coagulation also worked well for ^{144}Ce removal (>90%) but less so for ^{85}Sr removal (~70%). As expected, none of the coagulants were effective for ^{137}Cs removal. Based on experience at other sites, polyvalent isotopes such as ^{60}Co , $^{95}\text{ZrNb}$, rare earths, and actinides should scavenge well during the precipitation-coagulation process. Coagulation at high pH (10.5 to 11.5) is favored since better radionuclide removals are generally obtained at these levels.

Cesium removal may be increased in the coagulation process through the addition of grundite clay. Reported dfs for cesium in ORNL waste treatment studies with 0.7 lb of grundite clay added per 1000 gallons were 21 in the laboratory and 8 in the pilot plant.⁽¹¹⁾ Although these dfs may be sufficient for 200-West laundry waste, the use of a cesium-selective exchanger bed should achieve higher dfs with less solid waste generated.

PROCESS SELECTION RATIONALE

Chemical coagulation-flocculation and sedimentation processes were selected for bench-scale studies because they are widely used throughout the nuclear industry both in this country and abroad for the treatment of low and intermediate level wastewaters. The insolubility of the hydroxides, carbonates, and phosphates of many cationic fission products allows chemical precipitation to achieve at least a partial removal of activity, particularly if coagulants and carriers are added to give bulk to the precipitate. The main advantages of treatment by coagulation-flocculation and sedimentation are low cost, the ability to handle a wide range of inactive constituents in the liquid and the use of equipment and procedures for which considerable experience exists. It is anticipated that this treatment, perhaps coupled with ion exchange polishing, will achieve the objective of reducing the radioactivity of the laundry wastewater to a level as low as technically and economically practical. In addition, sediment radioactivity levels in the U-14 Ditch would be expected to decrease as a result of this treatment.

PILOT PLANT STUDIES

Bench-scale pilot plant studies have been made in order to define the most effective methods for treating 200-West laundry wastewater. The pilot plant flowsheet used to evaluate chemical coagulation and clarification of laundry wastewater is illustrated in Figure 1 with the principal components of the pilot plant listed in Table 9.

PILOT PLANT DESCRIPTION

The pilot plant was operated at a flow rate of 0.5 gal/min, and the first step was to pump raw wastewater to the chemical addition tank where the chemical coagulants were added and mixed in. Sodium hydroxide was also added at this point to increase the pH of the mixture when Fe^{+3} was used as a coagulant. The wastewater then overflowed to the rapid mix tank where polyelectrolyte was added to aid in flocculation. The treated wastewater overflowed the rapid mix tank to the flocculation tank which was gently mixed with a paddle stirrer rotating at 6 rpm to promote growth of the floc.

Separation of the floc from the liquid occurred in the tube settler where the clarified liquid overflowed a wier to the filter and the sludge was collected in the conical bottom of the clarifier. The settling tubes also aided in flocculation and promoted more rapid sedimentation than is normally possible in an open clarifier. Sludge was collected in the bottom of the clarifier and periodically drawn off and transferred to a sludge storage tank.

Filtration was accomplished with a multi-media or mixed media filter containing coarse grade anthracite coal, medium grade silica sand, and fine grade garnet sand. When backwashed to removed filtered material the bed settled with the coarse coal on top and the fine garnet sand on the bottom. No distinct bands of filter media were present since some intermixing of the three media occurred. However, filtration occurred in the direction of coarse media to fine media which permitted greater utilization of the filter media void space and longer filter runs than with conventional single media beds. (Single media beds tend to plug rapidly at the surface where the fine media particles are located.)

FIGURE 1. Pilot Plant Flow Schematic

TABLE 9. Pilot Plant Components

1. Chemical addition tank; circular, 14 inches diameter by 24 inches high; equipped with a rapid mix stirrer. Capacity is 13 gallons.
2. Rapid mix tank; rectangular, 8.5 inches by 11.5 inches and 13 inches deep. Equipped with a rapid mix stirrer. Capacity is 4 gallons.
3. Flocculation tank; circular, 18 inches diameter by 30 inches high, equipped with a slow speed paddle stirrer. Capacity is 28 gallons.
4. Tube settler with hopper bottom; 12 inches by 18 inches in plan view; tubes are 9 inches long inclined 60° to the horizontal, hexagonal shape, 1-1/2 inches by 1 inch in the long and short dimensions.
5. Multimedia filter; 12 inches by 12 inches surface area; 4 inches 10 x 40 mesh anthracite coal; 2 inches of 20 x 40 mesh silica sand; and 2 inches of 40 x 80 mesh garnet sand on a 3 inch bed of gravel. Equipped with a three tube header on the effluent line.
6. Sludge storage tank; rectangular, 18 inches square at the top by 25 inches deep; 34 gallon capacity.
7. Ion exchange feed tank; rectangular, 72 inches by 30 inches and 36 inches deep; 340 gallon capacity.
8. Ion exchange column; glass tube one inch diameter and 30 inches high filled to a depth of 24 inches with Na^+ based 20 x 50 mesh Buckhorn, New Mexico clinoptilolite.
9. Raw waste feed pump; centrifugal variable speed, 0.6 gpm capacity.
10. Filter effluent pump; centrifugal, constant speed, 5 gpm capacity; liquid level control.
11. Sludge pump; centrifugal, constant speed, 5 gpm capacity; manual control.
12. Ion exchange feed pump; peristaltic circular tube pump; variable speed.

The filtered effluent was then pumped to the ion exchange feed tank where it is stored for use in the ion exchange studies. Commercially available Buckhorn, New Mexico clinoptilolite was used in the ion exchange column to remove cesium, strontium and other radionuclides not removed by the chemical coagulation step. The zeolite was converted to the Na^+ form by regeneration with 10 bed volumes of 3% sodium chloride solution. Regeneration of the zeolite was performed to remove strontium impurities which would interfere with determination of the cold tracer strontium decontamination factor. This regeneration step would not be needed in actual practice where the zeolite is to be discarded.

Samples of wastewater for the pilot plant studies were collected from the sump located at the northeast corner of the 2724-W Building. This sump collects wastewater from the non-regulated washer as well as the regulated washer and mask cleaning facility. Approximately 20% of the total flow is from the non-regulated washers.

The wastewater was pumped to a 2500-gallon storage tank where it was sampled for total beta and total alpha analysis. Since the pilot plant equipment is located in the non-regulated zone of the Physical Sciences Laboratory at Battelle, the wastewater used in the pilot plant was limited to a total beta and total alpha concentration of 10 pCi/ml. The sample analysis was completed within several hours of the transfer of the wastewater to the storage tank to permit transport of the wastewater to the laboratory in the same day. The wastewater was transported to the laboratory in 200-gallon hard rubber containers. One out of five batches of wastewater collected in the storage tank was rejected because the total beta concentration exceeded the limit. Another batch was rejected because of a lack of time to complete the analysis the same day. Freezing weather prohibited storing the wastewater overnight.

About 1 mg/l of non-radioactive tracers, Cs^+ , Sr^{+2} and Eu^{+3} was added to the wastewater prior to processing through the pilot plant. These tracers permitted measurement of the cesium, strontium, and rare earth decontamination factors by atomic absorption or flame emission spectrophotometry. The Eu^{+3} is also a substitute for the actinides and other elements which are insoluble

under alkaline conditions. The tracer analysis was performed with a spectrophotometer using a graphite tube furnace for the Sr^{+2} , and Eu^{+3} analysis and flame emission for the Cs^{+} analysis. Chemical analyses were performed according to methods outline in the previous report on this project.

RESULTS AND DISCUSSION OF PILOT PLANT STUDIES

Coagulation, ion exchange, and sludge characteristics were evaluated and the results are reported in the following text. A waste treatment flow-sheet was also developed.

COAGULATION STUDIES

Pilot runs were made to test ferric hydroxide coagulation, clinoptilolite addition, and lime coagulation.

Ferric Hydroxide Coagulation

The use of ferric chloride as a primary coagulant along with magnesium chloride and calcium chloride at pH 11 was investigated in a series of pilot plant runs. The purpose of this test was to evaluate the effectiveness of this combination of coagulants under continuous flow conditions. Previous studies employing static or jar tests had disclosed this combination of coagulants to be the most effective of several coagulants tested for strontium and suspended solids removal. Coagulation with ferric hydroxide produces a very clear effluent, while the addition of Mg^{+2} and Ca^{+2} improves the settling and scavenging characteristics of the precipitate. Both Fe^{+3} and Mg^{+2} precipitate as gelatinous hydroxides at pH 11, whereas Ca^{+2} reacts with PO_4^{-3} and CO_3^{-2} in the wastewater to form insoluble $Ca_5(OH)(PO_4)_3$ and $CaCO_3$. The $Ca_5(OH)(PO_4)_3$ or calcium hydroxyapatite is formed preferentially to $CaCO_3$ as long as sufficient PO_4^{-3} is available. Calcium hydroxy apatite is an effective scavenger for strontium, rare earths, and transuranics.

Table 10 presents decontamination factors measured at three different Fe^{+3} dosages on the same batch of laundry wastewater which was traced with about 1 mg/l of Cs^+ , Sr^{+2} , and Eu^{+3} . The results showed no significant increase in the removal of Sr^{+2} , Eu^{+3} , and total beta at Fe^{+3} dosages above 21 mg/l. Cesium concentrations were essentially unaffected by this treatment. Phosphorus, suspended solids and turbidity in the filter effluents at the different Fe^{+3} dosages were not substantially different and averaged about 5 mg/l, 4 mg/l and 0.4 NTU, respectively. These and other data are given in the Appendix.

TABLE 10. Decontamination Factors Measured at Different Fe^{+3} Dosages

Run No.	Fe^{+3} Dosage, mg/l	Average Decontamination Factor ^a		
		Strontium	Europium	Total Beta
1	21	2.0	>92	1.2
2	42	2.2	>92	1.2
3	63	2.0	>92	1.1

Other Chemicals Added: 20 mg/l Mg^{+2} as MgCl_2 , 40 mg/l Ca^{+2} as CaCl_2 , NaOH to pH 11 and 1 mg/l Magnifloc 836-A.

^aDecontamination factor is equal to the influent concentration divided by the filter effluent concentration.

Three different batches of laundry wastewater were collected for testing to determine possible differences in processing. Decontamination factors for Sr^{+2} , Eu^{+3} , and total beta are given in Table 11. The third batch of wastewater collected on January 6, 1977, required five times as much Fe^{+3} to effectively coagulate and flocculate the suspended solids. Even at the higher level of Fe^{+3} dosage the turbidity of the filter effluent was about twice that of the filter effluents from the first two batches. In view of the higher Na^+ and phosphorus concentrations in the third batch of wastewater it was suspected that a higher ratio of washwater to rinsewater was present compared to the first two batches of wastewater. The Sr^{+2} df for the third batch was greater than the first two; however, the df for Eu^{+3} was less. Total beta dfs remained low in all cases. A major fraction of the total beta observed in the first batch (11-30-76) was found to be ^{137}Cs which was not removed by the coagulation step. It is believed that ^{137}Cs may have been present in the third batch since treatment of this waste through a clinoptilolite column, without chemical clarification, reduced the total beta below detection limits.

TABLE 11. Decontamination Factor for Different Batches of Laundry Wastewater

<u>Run No.</u>	<u>Date Sampled</u>	<u>Fe⁺³ Dosage</u>	<u>Average Decontamination Factor</u>		
			<u>Strontium</u>	<u>Europium</u>	<u>Total Beta</u>
1	11-30-76	21	2	>92	1.2
4	12-8-76	20	1.7	>93	2.2
5	1-6-76	100	5.9	33	1.3

Other Chemicals Added: 20 mg/l Mg⁺² as MgCl₂, 40 mg/l Ca⁺² as CaCl₂, NaOH to pH 11 and 1 mg/l Magnifloc 836-A.

Clinoptilolite Addition

The addition of fine particulate clinoptilolite to laundry wastewater during chemical coagulation with Fe⁺³, Mg⁺², and Ca⁺² salts was evaluated as a means of reducing the cesium and strontium concentrations at this step. Two runs were made with untreated -50 mesh clinoptilolite at 0.5 and 1.0 lb per 1000 gallons and two runs with treated -50 mesh clinoptilolite at 1.3 and 1.7 lb per 1000 gallons. Decontamination factors for these runs are listed in Table 12. Treatment of the clinoptilolite consisted of dilute HCl addition to a slurry of the -50 mesh zeolite until pH 2 was maintained for 5 min. The zeolite was then treated with saturated NaCl to convert it to the Na⁺ form. The acid and NaCl treatment serves to remove any strontium ions or carbonate minerals which may be present in the clinoptilolite.

The results in Table 12 reveal no significant increase in strontium dfs over previous runs made without clinoptilolite addition. Cesium df increased to a level of 1.5 at the highest addition of -50 mesh clinoptilolite. The cesium selectivity of clinoptilolite was much greater than that for strontium. For example, clinoptilolite from Hector, California, was about 14 times as selective for trace Cs⁺ than for trace Sr⁺² in the presence of Na⁺.

TABLE 12. Decontamination Factors Measures for
Coagulation with Clinoptilolite Addition

Run No.	Clinoptilolite Added, lbs/1000 gal	Average Decontamination Factors		
		Strontium	Cesium	Europium
6	0.5	2.2	1.0	>98
7	1.0	2.1	1.0	>98
8	1.3 ^a	1.5	1.2	>80
9	1.7 ^a	2.6	1.5	>80

^aClinoptilolite treated with dilute HCl and saturated NaCl solution.

Chemicals Added: Fe^{+3} 20 mg/l, Mg^{+2} 20 mg/l, Ca^{+2} 40 mg/l, NaOH to pH 11; and magnifloc 836-AI 1 mg/l.

Lime Coagulation

A series of four pilot plant runs were made to evaluate lime as a coagulant in a continuous flow system. Three of the runs involved variable polyelectrolyte addition to establish an optimum level for a rapid settling flow. The fourth run was carried out at the optimum polyelectrolyte addition over a sufficient time period to collect sludge for volume measurements. Decontamination factors obtained by lime coagulation are presented in Table 13. The relatively high dfs measured for strontium and low dfs for europium in Runs 10-12 cannot be explained. Only one filter effluent sample was available for Run 10. It is believed that the dfs for Run 13 are more representative of lime coagulation than the results of Runs 10-12.

ION EXCHANGE

Approximately 200 gallons of the first batch of laundry wastewater was treated by coagulation with 20 mg/l Fe^{+3} , 20 mg/l Mg^{+2} , 40 mg/l Ca^{+2} , and 1 mg/l 836-A polymer at pH 11 to obtain sufficient feed for the ion exchange study. The ion exchange column consisted of a 1-in. wide diameter glass pipe filled to a depth of 24 in. with 20 x 50 mesh clinoptilolite. The feed

TABLE 13. Decontamination Factors Obtained
with Lime Coagulation

Run No.	Polyelectrolyte Dosage, mg/l	Lime Dosage mg/l	Average Decontamination Factors		
			Strontium	Cesium	Europium
10	0.3	235	50	1.1	3
11	0.6	235	4.3	1.4	17
12	1.1	235	3.5	1.1	33
13	0.6	265	2.3	1.4	>100

*Different tank of waterwater from the same batch.

was pumped downflow through the clinoptilolite bed at a rate of 15 bed volumes per hour. Cesium and strontium breakthrough curves are illustrated in Figure 2. Decontamination factors exceeded 100 for Cs^+ and 34 for Sr^{+2} through 1700 bed volumes of effluent. The total df for strontium including a df of 2 through the coagulation step is 68. The run was terminated after 1870 bed volumes when filterable matter plugged the bed. The data indicated that both Cs^+ and Sr^{+2} breakthrough occurred at approximately 1500 bed volumes.

It was anticipated that the clinoptilolite would also remove calcium and that strontium breakthrough would occur with calcium breakthrough or shortly thereafter. This did not prove to be the case as calcium breakthrough occurred within 100 bed volumes. The breakthrough data indicated that Buckhorn, New Mexico clinoptilolite is much more selective for strontium in the presence of Na^+ and Ca^{+2} than Hector, California clinoptilolite. Further studies on basic ion exchange characteristics of Buckhorn clinoptilolite would be required to establish its strontium selectivity. Efforts to correlate strontium breakthrough with hardness breakthrough as a process control measure were therefore abandoned. Possibly, a sensitive radiation detection device can be placed next to an ion exchange column to signal radionuclide breakthrough. The concentration of radionuclides in the feed stream will, in most cases, be too low for successful in-stream monitoring to determine breakthrough.

FIGURE 2. Cs^+ and Sr^{+2} Breakthrough Curves.
FLOW: 15 COLUMN VOLUMES/HR
COLUMN DIMENSIONS: 1 INCH DIAMETER, 24 INCHES

About 200 gallons of the second batch of laundry waste received on 12-9-76 was treated similarly to the first batch to coagulate and remove suspended solids. Operating conditions for the clinoptilolite column were also the same as those for the first run. However, the column plugged within 17 hr. The ion exchange column was equipped with rubber stoppers reinforced by taping to the glass pipe and could not withstand pressures exceeding 15 psig.

A fresh column of clinoptilolite was prepared and placed in service with a 2-in. layer of coarse anthracite coal (10 x 20 mesh) on top of the ion exchange bed to prevent plugging at the surface of the bed. The feed stream was also treated with sodium hypochlorite to prevent biological growth from fouling and plugging the bed. Unfortunately, the hypochlorite apparently oxidized some of the ammonia in the waste to nitrogen which caused gassing in the bed. Poor contact of the feed stream with the clinoptilolite under these conditions resulted in lower dfs compared to the first column run.

Inspection of the top of the plugged clinoptilolite column disclosed that a hard crust had formed and some white material had settled in the tubing. X-ray diffraction analysis of both the white material and material separated from the crust revealed the presence of calcite or calcium carbonate. It is apparent that post-precipitation of calcium carbonate was occurring with this particular batch of laundry waste after coagulation and filtration. Plugging from calcite formation can be prevented by reducing the alkalinity of the feed stream to the clinoptilolite column. However, neutralization below pH 10 will result in NH_4^+ formation which will reduce the strontium capacity of the clinoptilolite to some extent. It may be necessary to adjust the pH of the ion exchange effluent in the range 6-9 in any case before discharge to U Pond. The acid requirement to accomplish neutralization to pH 7 is approximately 7.3 meq/l or about 3 lb of H_2SO_4 per 1000 gallons.

Approximately 200 gallons of the third batch of laundry wastewater collected on 1-6-77 was filtered through the mixed media filter without chemical coagulation. Suspended solids were reduced from 118 mg/l in the filter influent to 89 mg/l in the filter effluent. The filter effluent was then

FIGURE 3. Cs^{+} and Sr^{+2} Breakthrough Curves
for Clarified Batch No. 2 Wastewater.
FLOW: 15 COLUMN VOLUMES/HR
COLUMN DIMENSIONS: 1 INCH DIAMETER x 24 INCHES

pumped through a clinoptilolite column to evaluate the plugging characteristics of the bed with uncoagulated filtered waste. The pressure across the column increased from 0.5 psig to 5.5 psig over a period of 5.5 hr at which time the flow was reversed with distilled water for a few minutes to break any crust that might have formed on the surface. The bed was lifted only 2 in. during this time. There was no crust observed on top of the bed. However, the clinoptilolite granules appeared to be sticking together slightly. The ion exchange column reduced the suspended solids in the wastewater from 89 mg/l to 25-30 mg/l. The flow reversal was not very effective for reducing the pressure across the bed since the bed was restarted at a pressure of 4.5 psig. Flow to the column was continued for 9 more hours at which time the pressure had increased to 10.6 psig. The flow was again reversed with distilled water, but the zeolite granules could not be readily fluidized because they agglomerated. Furthermore, the bed appeared to be plugged in depth rather than at the surface. The run was terminated at this point.

A portion of the third batch of laundry wastewater was also coagulated and filtered (see Run No. 5, Table 3) to prepare feed for another ion exchange run through a 1-in. diameter, 24-in. deep bed of clinoptilolite. Substantially greater caustic addition was required during coagulation since more FeCl_3 had to be added to clarify this batch of wastewater. Cesium and strontium dfs were found to be much lower than in previous ion exchange runs. Investigation subsequently revealed that a 5-lb bottle labeled reagent-grade NaOH, used to prepare the caustic solution for pH adjustment, actually contained KOH. This introduced over 200 mg/l K^+ to the ion exchange influent which caused immediate Sr^{+2} breakthrough and interfered with the analysis for Cs^+ . The K^+ is also much more competitive than Na^+ with respect to uptake of Sr^+ . A small ion exchange column was prepared to determine Cs^+ breakthrough using ^{137}Cs tracer. The breakthrough of ^{137}Cs is illustrated in Figure 4. Decontamination factors exceeding 100 were achieved with the column for about 350 bed volumes after which ^{137}Cs began to breakthrough.

FIGURE 4. Cesium Breakthrough Curve for Small Clinoptilolite Column with K^+ Contaminated Feed.
FLOW: 31 COLUMN VOLUMES/HR
COLUMN DIMENSIONS: 0.6 cm DIAMETER x 20 cm HEIGHT

SLUDGE CHARACTERISTICS

Sludges were collected in the clarifier during the coagulation steps and subsequently transferred to the sludge storage tank where the settled sludge volume was measured over a period of time. Figures 5 through 7 illustrate settled sludge volumes as a function of time. The volume of settled sludge is expressed as a percentage of the volume of wastewater processed through the clarifier. All sludges, except that from the third batch of laundry wastewater, settled to 4 to 5% of the total wastewater volume within 3 hr.

Gravity thickening of settled sludge was evaluated by placing a liter of mixed sludge from the storage tank in a graduated cylinder tank and allowing it to settle further. Supernatant liquid had previously been decanted from the storage tank prior to removing the settling sludge. Figure 8 presents settled sludge volumes as a function of time for sludges which had settled at least 20 hrs in the storage tank. Additional sludge volume reductions are possible by gravity thickening, although thickening of sludge from high Fe^{+3} dosage on Batch 3 appeared to be quite slow.

Filtration and centrifugation of settled sludge samples from the sludge storage tank was evaluated. The filtration tests were conducted by filtering 200 ml sludge through a 12.5 cm diameter filter paper on a Buchner funnel. The filtrate was collected in a 100 ml graduated cylinder inside a filter flask. A vacuum of 12 in. of mercury was placed on the filter flask as the sludge sample was transferred to the Buchner funnel.

Sludge samples from Runs 1-3 required only 60 to 75 sec for 100 ml of filtrate to pass through the filter. The wet sludge was readily dewatered to damp cake on the filter paper. The filter cakes were removed from the filter when cracks began to appear in the cakes. Moisture content varied between 9 and 11%.

Centrifugation tests were conducted by placing 100 cm³ of settled sludge in polyethylene tubes and centrifuging at 200 g for designated periods of time. The volume of sludge after centrifugation is given in Table 14. Sludge from the storage tank contained approximately 1% dry weight solids.

FIGURE 5. Settled Sludge Volume.

FIGURE 6. Settled Sludge Volume.

FIGURE 7. Settled Sludge Volume.

FIGURE 8. Thickened Sludge Volume vs Time.

TABLE 14. Centrifuged Sludge Volumes

<u>Sludge</u>	<u>Time, Min.</u>	<u>Sludge Volume, cm³</u>	<u>Percent Dry Solids</u>
Run 2	1	26	4.3
Run 2	2	23	4.4
Run 2	4	20	-
Run 1	2	19	4.0
Run 3	2	18	4.3

On the basis of the results obtained on the centrifugation and filtration tests, a rotary vacuum filter would be superior to a continuous centrifuge to minimize the moisture content of the dewatered sludge. It is anticipated that the sludge cake from a vacuum filter could be packaged in a plastic lined container and transferred directly to a burial ground without drying.

RECOMMENDED FLOWSHEET

The processing steps recommended for treatment of the 2724-W laundry wastewater are illustrated in the flowsheet given in Figure 9. The laundry waste is initially routed through a grit chamber to remove heavy particulates such as sand and gravel. The wastewater is then routed through a debris screen which removes large suspended or floating material such as cloth, string and plastic items. A retention basin is provided for surge capacity and equalization. Wastewater is pumped from the basin to a rapid mix tank where chemicals are added to scavenge radionuclides and coagulate suspended solids. Coagulant chemicals recommended for this step include ferric chloride, calcium chloride, and magnesium chloride or sulfate. Magnesium sulfate is normally less expensive than magnesium chloride; however, sulfate salts cannot be premixed with calcium salts since calcium sulfate precipitates. Separate chemical feed tanks for each of the coagulant chemicals may be desirable to allow adjustment of the individual chemical dosages to fit process needs. Sodium hydroxide and polyelectrolyte are also added at this point to maintain the pH at 11 and flocculate the coagulated solids.

The coagulated wastewater then flows to a flocculator where it is gently agitated to allow particulate growth. Residence time in the flocculator is generally 20 min or more; subsequently, the clarifier receives the flocculated wastewater for sedimentation of the particulate matter. Several different types of clarifiers are commercially available for this purpose. Clarifiers equipped for continuous sludge removal are recommended in favor of the slightly inclined tube settler considering the relatively large volume of sludge produced (5-10% of total flow). Slightly inclined tubes would fill rapidly with sludge and would require frequent draining to remove the sludge. Frequent draining of the tubes substantially increases the amount of recycled wastewater from these units.

Wastewater overflowing the clarifier is next routed to a mixed media filter to remove residual particulate matter. Sludge is pumped from the clarifier to a storage tank where gravity thickening occurs. Excess water removed by decantation is recycled back to the retention basin. The thickened sludge is pumped to a rotary vacuum filter where most of the unbound water is removed. The sludge cake is removed from the filter, packaged, and transferred to a burial ground. The filtrate stream is also recycled to the retention basin.

Clarified wastewater flows to the mixed media filter until a predetermined headloss is reached at which time the filter is backwashed to remove particulate matter from the media. Filter backwash water is recycled to the retention basin.

The filtered wastewater flows to a backwash storage tank and from there is pumped through a series of ion exchange columns containing 20 x 50 mesh clinoptilolite for removal of cesium and strontium. Plugging in the zeolite column should be minimal with a short detention time in the filter backwash storage tank to reduce the risk of post-precipitation of CaCO_3 . The mixed media filter bed should have sufficient fine garnet media to assure effective particulate removal to avoid plugging. Four ion exchange columns are recommended which allows at least two in service during operational periods. One of the columns may be undergoing discharge of spent zeolite and refilling while the fourth column is being held in reserve. The fresh zeolite columns

are placed at the end of the series as the exhausted columns are removed from the head end. Maximum flow through each of the columns should not exceed 0.25 gpm/ft^3 of zeolite. The ion exchange treatment unit is equipped with a zeolite slurry tank for transferring fresh zeolite to the column. Spent zeolite is backwashed from the column to the rotary vacuum filter.

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TABLE A-1. Iron Coagulation of Laundry Wastes at Different Fe⁺³ Dosages

No.	Fe ⁺³ Dosage, mg/l	Suspended Solids, mg/l		P, mg/l		Ca, mg/l		Mg, mg/l		Na, mg/l		Eu, mg/l		Sr, mg/l		Cs, mg/l		Turbidity, Relative Units					
		Inf.	F.E.	Inf.	F.E.	Inf.	F.E.	Inf.	F.E.	Inf.	F.E.	Inf.	F.E.	Inf.	F.E.	Inf.	F.E.	Inf.	F.E.				
1	21	72	5.0	0.7	53	5	25	23	0.54	0.28	430	430	.92	0.02	<0.01	1.15	0.81	1.13	1.19	1.01	44	0.9	0.4
		7.3	4.7	75	5	28	26	0.39	0.26	280	410	0.01	<0.01	0.93	0.57	1.13	1.07	1.1	0.6				
2	42	102	6	5	54	5	5.5	13	28	26	200	420	0.9	0.01	<0.01	1.15	0.74	1.13	1.17	1.12	38	1.5	0.5
		3	-	-	9	6.5	23	23	0.21	0.13	450	430	0.04	<0.01	0.74	0.53	1.25	1.20	1.0	0.3			
		4.9	1	8	5.5	24	24	0.21	0.13	440	430	0.01	<0.01	1.14	0.56	1.24	1.21	1.2	0.5				
3	63	102	1.2	5	54	8	4.5	13	24	23	200	420	0.92	0.04	<0.01	1.15	0.85	1.13	1.19	1.17	37	1.4	0.4
		2.0	5	4	3	24	24	0.35	0.13	460	400	0.02	<0.01	0.69	0.63	1.19	1.17	1.2	0.2				

Inf. - Influent, T.E. - Tube Settler Effluent, F.E. - Filter Effluent
Common Chemicals Added: Ca²⁺, 42 mg/l; Mg²⁺, 21 mg/l; 836A polymer, 1.0 mg/l; NaOH added to pH 11.0 to 11.7

TABLE A-2. Iron Coagulation of Different Batches of Laundry Waste

No.	Fe ⁺³ Dosage, mg/l	Suspended Solids, mg/l				P, mg/l		Ca, mg/l		Mg, mg/l		Na, mg/l		Eu, mg/l		Sr, mg/l		Cs, mg/l		Turbidity, Relative Units						
		Inf.	T.E.	F.E.	Inf.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	Inf.	T.E.					
1	21	72	5.0	0.7	53	5	5	25	23	0.54	0.20	430	430	.92	0.02	<0.01	1.15	0.81	1.19	1.01	44	0.9	0.4			
		7.3	4.7		75	5	5	28	26	0.39	0.26	280	410	0.01	<0.01	0.93	0.57	1.13	1.07			1.1	0.6			
4	20	66	29.5	2.4	65	8	6	12	31	28	3.36	2.14	108	303	280	0.93	<0.02	<0.01	1.22	0.68	1.16	1.05	0.89	26	2	0.3
		14	0.4		8.5	7.5	34	30	34	30	0.48	0.38	308	308	293	<0.01	<0.01	0.84	0.72	1.16	1.09	0.9	0.3			
		14	0.4		8	8	33	32	33	32	0.18	0.09	370	370	392	0.01	0.01	0.81	0.85	1.18	1.16	0.8	0.3			
5	100	111	19	8.7	107	27	28	20	24	24	4.2	4.9	307	410	420	1.0	0.03	0.03	1.53	0.39	1.40			55	4	1.2
		14	3.3		26	24	24	23	24	23	2.7	3.2	332	332	355	0.04	0.03	0.33	0.31			4	1.2			

Chemicals Added: Ca²⁺, 40 mg/l; Mg²⁺, 20 mg/l; 836A Polymer, 1 mg/l; NaOH to pH 11.0; 1 mg/l each of Eu³⁺, Sr²⁺, Cs⁺

TABLE A-3. Results of Adding Clinoptilolite to Iron Coagulation of Laundry Waste

No.	Fe ⁺³ Dosage, mg/l	Suspended Solids,			P, mg/l			Ca, mg/l			Mg, mg/l			Mn, mg/l			Eu, mg/l			Sr, mg/l			Cs, mg/l			Turbidity, Relative Units		
		Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.	Inf.	T.E.	F.E.
6	0.5	72	1.0	1.0	53	8.5	5.5	12	28	24	2.4	0.21	0.16	130	360	370	0.98	0.04	<0.01	1.34	0.88	0.63	0.98	1.08	1.04	44	1.3	0.4
			1.0	1.0		10	6.5	29	26			0.24	0.14		350	360		0.02	<0.01		0.78	0.61	1.07	1.05		1.1	0.4	
7	1.0	72		4.0	53			12	32	27	2.4	0.17	0.12	130	410	410	0.98	<0.01	<0.01	1.34	0.96	0.61	0.98	0.92	1.00	44	0.8	0.4
				8.7		8	5		33	26		0.20	0.11		390	380		<0.01	<0.01		1.03	0.65	0.84	0.99		1.0	0.4	
8	1.3*	66	5.3	1.3	68	14	9	12	35	35	2.9	0.7	0.6	49	240	327	0.8	<0.01	<0.01	1.09	0.96	0.61	1.03	0.83	0.83	25	1.0	0.3
			2.0	0		12.5	11		35	31		0.6	0.5		408	224		0.01	<0.01		1.02	0.72	0.85	0.85		1.1	0.4	
			0.7	0		13.5	9		38	33		0.5	0.4		210	241		0.02	<0.01		1.07	0.75	0.85	0.83		0.8	0.4	
			3.3	2.0		14	8.5		35	33		0.5	0.4		325	210		<0.01	<0.01		0.88	0.74	0.86	0.86		1.0	0.4	
9	1.7*	66	5.3	0	68	10.5	6.5	12	35	20	2.9	2.6	2.0	49	218	113	0.8	<0.01	<0.01	1.09	0.98	0.37	1.03	0.96	0.55	25	2.0	0.5
			4.7	2.0		11.0	7.0		38	30		2.0	1.4		268	176		0.01	<0.01		0.83	0.48	0.95	0.80		24	1.1	0.5

*Clinoptilolite treated with dilute HCl and saturated NaCl.
Common Chemicals Added: Ca²⁺, 21 mg/l; Mg²⁺, 21 mg/l; 836A polymer,
1.0 mg/l; Fe³⁺, 21 mg/l; NaOH to pH 11; 1 mg/l
each Eu³⁺, Sr²⁺, Cs⁺

TABLE A-4. Lime Coagulation of Laundry Waste

No.	Polyelectrolyte mg/l	Suspended Solids, mg/l			P, mg/l			Ca, mg/l			Mg, mg/l			Mn, mg/l			Eu, mg/l			Sr, mg/l			Cs, mg/l			Turbidity, Relative Units		
		Inf.	I.E.	F.E.	Inf.	I.E.	F.E.	Inf.	I.E.	F.E.	Inf.	I.E.	F.E.	Inf.	I.E.	F.E.	Inf.	I.E.	F.E.	Inf.	I.E.	F.E.	Inf.	I.E.	F.E.	Inf.	I.E.	F.E.
10	0.3	66	45	7.3	52	18.5	16	10	58	43	2.2	11.6	2.7	140	160	150	1.00	0.60	0.32	1.12	0.43	0.02	0.91	0.89	0.86	47	36	3
11	0.6	66	44			20	10.5	54	36			10.2	1.7	170	120		0.36	0.07		0.74	0.40		0.93	0.86		27	3	
			40			12.5	7.5	62	36			9.3	5.5	150	80		0.18	0.05		0.58	0.12		0.82	0.45		20	3	
12	1.2	66	50			18.5	10	55	42			9.7	2.7	170	170		0.26	0.05		0.66	0.32		0.91	0.74		24	6	
			32			16	7.5	52	44			9.5	1.1	170	170		0.31	0.02		0.84	0.20		0.93	0.88		21	2	
			20	2.7		18.5	6	56	40			9.5	1.0	180	180		0.22	0.02		0.58	0.45		0.91	0.85		48	21	1

Chemicals Added: Ca(OH)₂, 231 mg/l; Mg²⁺, 20 mg/l; pH, 11.0

Chemicals Added: Ca(OH)₂, 231 mg/l; Mg²⁺, 20 mg/l; pH, 11.0

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