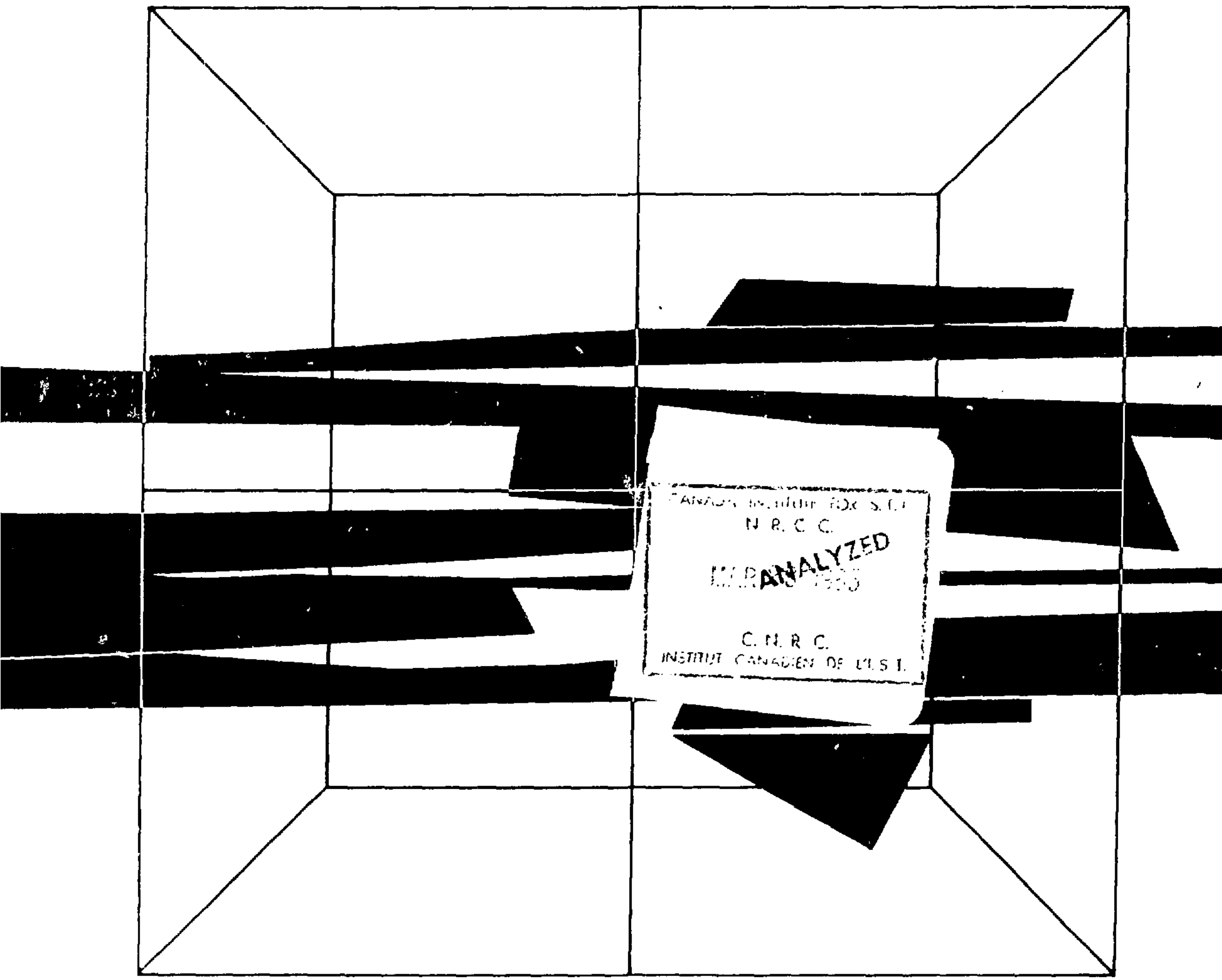
Removal of Radium-226 from Uranium Mining Effluents

Final Report of a Joint Government-Industry Program

Report EPS 3/HA/3
December 1984



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REMOVAL OF RADIUM-226 FROM URANIUM MINING EFFLUENTS

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by

D.W. Averill
D. Moffett
R.T. Webber
L. Whittle
J.A. Wood

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PREFACE

An intensive process development program for the removal of radium-226 from uranium mining effluents was undertaken jointly by representatives of the Government of Canada and the Canadian Uranium Mining Industry between 1978 and 1980. One of the implicit objectives was the dissemination of the information developed. To ensure complete documentation of the program, this information has been compiled in three volumes: Volume I summarizes the results of the program; Volume II provides the complete text; and Volume III consists of appendices. Volume I is reproduced in this report. Volumes II and III may be obtained from:

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PRÉFACE

Un programme intensif de mise au point d'un procédé d'élimination du radium-226 des effluents des mines d'uranium a été entrepris par des représentants du gouvernement canadien et des entreprises exploitant l'uranium au Canada entre 1978 et 1980. L'un des ojectifs implicites était la diffusion des résultats obtenus. Pour documenter complètement le programme de recherche, trois volumes ont été préparés: le Volume I résume les résultats du programme; le Volume II contient le texte complet; le Volume III rassemble toutes les annexes. Le présent rapport est le Volume I. On peut se procurer les Volumes II et III en s'adressant à:

Environnement Canada
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C.P. 5050
Burlington (Ontario)
L7R 4A6

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1 INTRODUCTION

Uranium mining and milling operations usually generate large quantities of solid and liquid waste materials. A slurry, consisting of waste rock and chemical solutions from the milling operation, is discharged to impoundment areas (tailings basins). The solids settle out in these basins leaving a clear supernatant which contains a variety of dissolved materials. In temperate climates, where precipitation exceeds evaporation, it is necessary to discharge this liquid effluent to local watercourses.

Most of the radioactive material dissolved in tailings slurries is precipitated by the addition of lime and limestone prior to discharge from the mill. However, the activity of one radioisotope, radium-226, remains relatively high in the tailings basin effluents. Radium-226 is of particular environmental concern because of its radiotoxicity to humans. All uranium mining and milling liquid effluents must be treated to remove radium-226 before the effluent is released to the environment.

In Canada, radium-226 is removed from uranium mining and milling effluents by the addition of barium chloride to precipitate barium-radium sulphate [(Ba, Ra)SO_4]. This process, which has been in use since 1965, generally consists of mixing a barium chloride solution with tailings basin effluent in a pipe or open channel, followed by solid/liquid separation in large sedimentation ponds. Coagulants such as ferric or ferrous sulphate may be used to improve sedimentation efficiency. Although dissolved radium-226 activities are generally reduced effectively, the process is considered to have two undesirable characteristics: the first related to suspended radium-226 in the effluents and the second to ultimate disposal of the (Ba, Ra)SO_4 sludge. Barium-radium sulphate is a fine, slow settling crystalline solid which is not easily removed in simple sedimentation systems. Depending on the size of the sedimentation ponds, hydrological factors, and whether or not the mill uses coagulants as part of the treatment process, appreciable quantities of particulate radium-226 may be present in the pond effluents. The second area of concern is the potential problems associated with recovery of the precipitate from the ponds for ultimate disposal (IAEA, 1976).

Under the auspices of Environment Canada, a government-industry mining task force established a radioactivity sub-group in 1974 to assist in the development of effluent guidelines and regulations for the uranium mining industry. Several recommendations for further research were identified (Radioactivity Sub-group, 1974). With respect to radium removal, investigation of more effective removal methods was recommended, including the development of mechanical treatment systems as alternatives to settling ponds. In response to these recommendations and the previously noted concerns, Environment Canada's Wastewater Technology Centre (WTC) initiated a bench scale study in March, 1976. The study was designed to assess the feasibility of using precipitation, coagulation, flocculation and sedimentation for the removal of radium-226. In 1977, the study was accelerated with financial assistance from the Atomic Energy Control Board. The results were favourable, with improved radium removals obtained in bench scale batch tests using barium chloride as the precipitant and either alum or ferric chloride as the coagulant (Wilkinson and Cohen, 1977).

Based on of the feasibility study and concerns expressed by industry and government agencies, a more comprehensive bench scale and pilot scale process development and demonstration program was formulated. This program was jointly managed and funded by government and industry as outlined in Table 1. The results of the joint study are summarized in this report.

TABLE 1 JOINT GOVERNMENT-INDUSTRY PROGRAM PARTICIPANTS

	Represented on		Contributing			
Participating Organization	Management Panel	Working Group	Financial Support	Pilot Plant Staff	Laboratory Services	Site Services
AMOK Limited	*		*			
Atomic Energy Control Board	*		*			
Denison Mines Limited	*	*	*	*	*	*
Eldorado Nuclear Limited	*	*	*	*	*	*
The Elliot Lake Centre						*
Energy, Mines and Resources Canada	*		*			
Environment Canada	*	*	*	*	*	*
Gulf Minerals Canada Limited	*					
Key Lake Mining Corporation	*		*			
Madawaska Mines Limited	*		*			
Rio Algom Limited	*	*	*	*	*	*

2 EXPERIMENTAL PROGRAM

The general objective of the joint government-industry program was to develop and demonstrate a physical/chemical process for the removal of radium-226 from uranium mill tailings basin effluents. Specifically, the work was directed toward the optimization of barium-radium coprecipitation, solid/liquid separation and sludge dewatering. The goals established for the joint program are presented in Table 2. Experimental work was initiated in January 1978. The program was divided into several phases and scheduled as shown in Figure 1.

TABLE 2 JOINT GOVERNMENT-INDUSTRY PROGRAM GOALS

1) To develop at pilot scale, a physical-chemical process to reduce the radium-226 content of uranium mining and milling effluents.
2) To demonstrate at pilot scale, a reasonably achievable level of radium-226 in the effluent, with target levels of 0.37 Bq/L (10 pCi/L) total radium-226 and 0.11 Bq/L (3 pCi/L) dissolved radium-226.
3) To establish a data base for the design of a full-scale treatment system.
4) To evaluate process alternatives for dewatering the sludge produced in the physical-chemical treatment process.
5) To establish a data base for the design of a full-scale sludge dewatering process.
6) To estimate costs for physical-chemical treatment and sludge dewatering for a full-scale system.

Two wastewater processes were investigated (Figure 2). Both processes incorporated barium-radium coprecipitation in series-connected stirred tank reactors. In one process, coprecipitation was followed by rapid mixing of a chemical coagulant, flocculation in series-connected mechanical flocculators and solid/liquid separation in a clarifier. In the second process, barium-radium coprecipitation was followed directly by solid/liquid separation using chemically aided granular media filtration.

2.1 Effluent Treatment

The tailings basin effluent used in this program was obtained from the Quirke Mine of Rio Algom Limited near Elliot Lake, Ontario. The wastewater characteristics are summarized in Table 3.

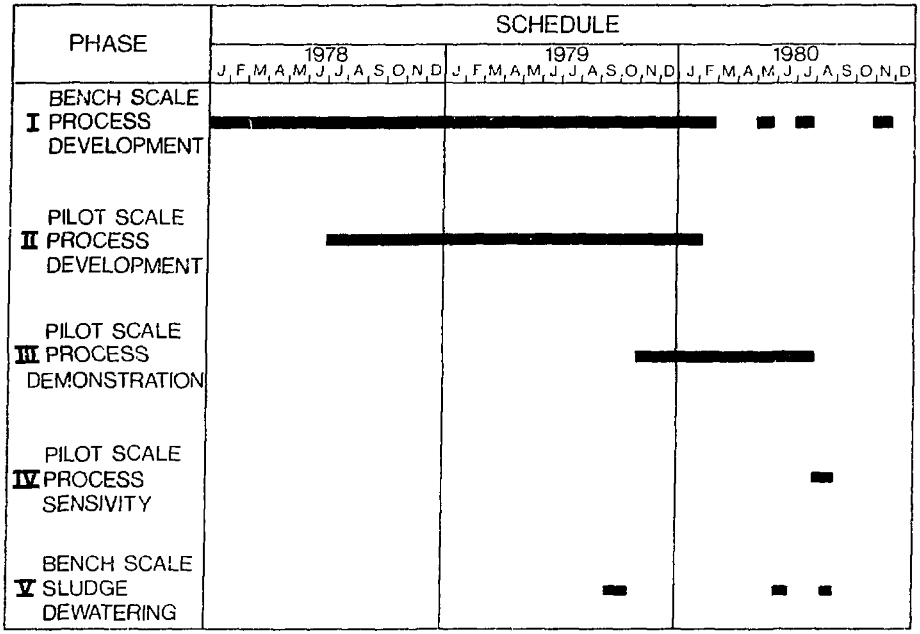


FIGURE 1 PROGRAM SCHEDULE

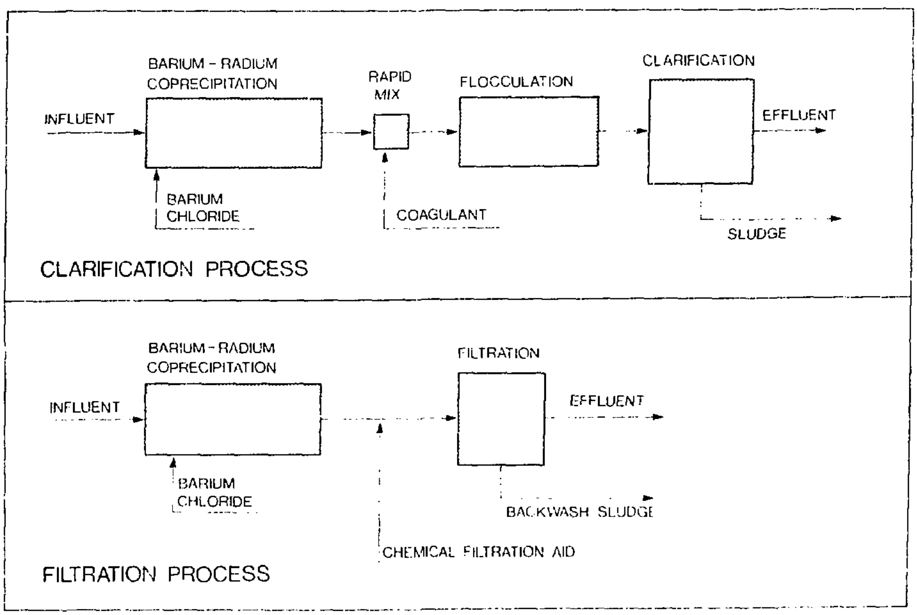


FIGURE 2 WASTEWATER TREATMENT PROCESSES

TABLE 3 TAILINGS BASIN EFFLUENT CHARACTERISTICS*

Parameter	Mean Value	Range
Total Radium-226 (Bq/L)	22.1	7.4 - 38.4
(pCi/L)	(597)	(200 - 1038)
Dissolved Radium-226** (Bq/L)	19.7	6.0 - 34.3
(pCi/L)	(532)	(163 - 927)
pH	9.3	6.7 - 10.6
Sulphate (mg/L)	1 187	490 - 1 870
Alkalinity (mg/L as $CaCO_3$)	108	27 - 200
Suspended Solids (mg/L)	≃ 2	< 2 - 36
Nitrate Nitrogen (mg/L)	66.5	0.2 - 154
Ammonia Nitrogen (mg/L)	23.5	6.9 - 41

* Batch samples received at the Wastewater Technology Centre, January 1978 to November 1979.

** Radium-226 activity in filtrate passing 0.45 μ m membrane filter.

Laboratory scale batch tests were used for the optimization of process variables. Bench scale continuous-flow tests were employed to confirm the batch test results. The pilot scale program was designed to confirm the results of the previous experiments, to identify scale-up relationships and to continue the optimization of process variables. The pilot scale program also included a demonstration phase during which the optimum operating conditions were tested under varying influent conditions for a period of several months.

The pilot plant was located below the main tailings basin dam of the Quirke Mine. The plant consisted of an enclosed 14 m highway trailer and an 11 m flatbed trailer which contained reactor and flocculator tanks, chemical feeding equipment and a circular clarifier. Additional, separate process units included a second circular clarifier, an inclined plate clarifier, and several granular media filters. The nominal capacity of the pilot plant was 23 L/min. Figure 3 is a plan view of the plant illustrating one of several process combinations tested during the program.

2.2 Sludge Treatment

The objectives of the bench scale sludge tests were to characterize the sludges produced by both the clarification and filtration processes and to assess sludge dewaterability. This portion of the work was conducted in cooperation with the Canada Centre for Mineral and Energy Technology (CANMET) of Energy, Mines and Resources

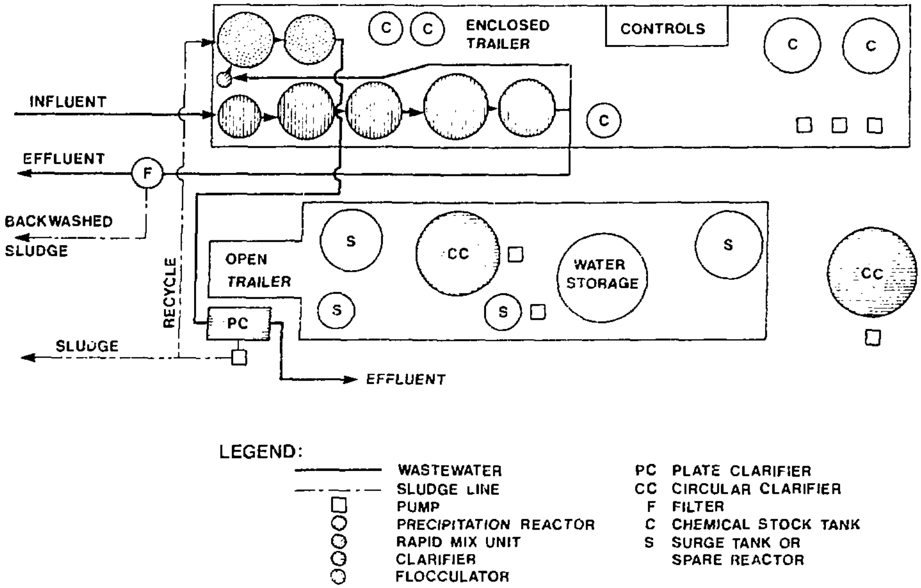


FIGURE 3 PLAN VIEW OF THE PILOT PLANT

Canada and also included sludges from other pilot scale and full-scale processes. The information obtained forms a component of the data base required to establish sludge disposal procedures. The results of the study are presented in a CANMET report (Skeaff and Campbell, 1982) and in Volume III, Appendix IV of the government-industry program report. Pilot scale sludge dewatering, originally planned as part of the overall program, was deferred pending identification of suitable sludge disposal options.

A second important topic, the mobility or the leachability of the radioactive constituents of the sludges was the subject of other studies (Bryant et al., 1979; IEC, 1980; Huck et al., 1982; Huck and Anderson, 1982). A third topic of interest was the feasibility of stabilization or chemical fixation of these sludges. A literature review of stabilization and fixation (solidification techniques) was also undertaken as part of this program and is included in Volume III, Appendix IV.

3 RESULTS

3.1 Barium-Radium Coprecipitation

The barium-radium coprecipitation operation was common to both the clarification and filtration processes. The objective of the operation was to reduce the dissolved radium-226 activity in the effluent of the tailings basin from an initial value of approximately 20 to 40 Bq/L (550 to 1100 pCi/L) to the project target of 0.11 Bq/L (3 pCi/L).

Batch tests were conducted in the laboratory to identify optimum process conditions and characterize the reaction kinetics. The coprecipitation operation was found to be sensitive to the barium chloride dosage and the reaction time; a barium dosage of 16 mg/L (29 mg/L as $BaCl_2 \cdot 2H_2O$) and a batch reaction time of 15 minutes were identified as optima. The reaction was apparently first order during the first 15 minutes, and the residual dissolved radium-226 activity was generally between 0.26 and 0.37 Bq/L (7 and 10 pCi/L). Subsequently, the dissolved radium-226 activity decreased very slowly.

The results of the batch experiments were used to calculate the required reactor volume and reactor configuration for continuous-flow tests. As a first approximation, alternative continuous-flow reactor systems were designed on the basis of first order kinetics, a 15 minute batch retention time, and a reduction of dissolved radium-226 activity from 37 to 0.11 Bq/L (1000 to 3 pCi/L). A reactor system consisting of two equal size series-connected continuous-flow stirred-tank reactors (CSTR's) with a total volumetric residence time of 90 minutes and a barium dosage of 16 mg/L was selected for testing. In 13 days of operation, the continuous-flow apparatus produced a mean effluent dissolved radium-226 activity of 0.37 Bq/L (10 pCi/L). The number of series-connected CSTR's and the total volumetric residence time were then increased in an effort to improve effluent quality. A reactor system consisting of three CSTR's with a total volumetric residence time of at least 113 minutes was required to produce a mean dissolved radium-226 activity of 0.19 Bq/L (5 pCi/L). The target of 0.11 Bq/L (3 pCi/L) was not achieved in these experiments.

During the pilot scale process development tests, the dissolved radium-226 activity in the effluent from the coprecipitation system was found to be highly variable (Figure 4). It was hypothesized that the variable performance of the system was related to several factors, including the operating conditions and seasonal variations in the

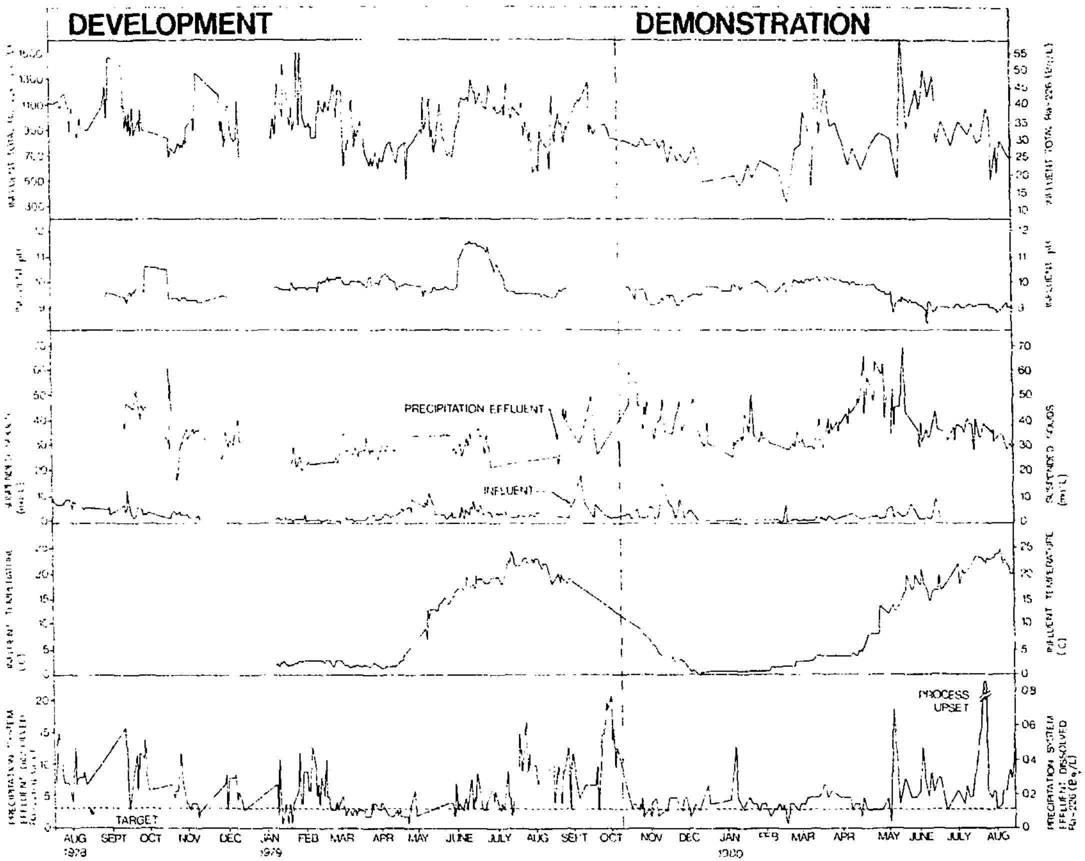


FIGURE 4 PERFORMANCE OF PILOT SCALE BARIUM-RADIUM COPRECIPITATION SYSTEM

wastewater temperature and/or chemical composition, but no single factor could be identified as the major source of the variations. Following the development tests, it was considered necessary to operate the coprecipitation system under steady state conditions during a demonstration period of approximately 12 months while monitoring the influent and effluent characteristics.

The reactor configuration selected for the process demonstration phase of the program consisted of five CSTR's in series with a total volumetric residence time of 70 minutes. That reactor system was required to satisfy the hydraulic demand of downstream units and provided the theoretical equivalent of the system originally selected on the basis of the bench scale test results (three CSTR's at 115 minutes). The demonstration phase was initiated in October, 1979 (Figure 4). The coprecipitation system initially performed very well, producing dissolved radium-226 activities which generally ranged between 0.11 and 0.19 Bq/L (3 and 5 pCi/L). An increase in the dissolved radium-226 activity was observed concurrently with an increase in the temperature of the tailings basin effluent. However, in mid-July, 1980, the performance of the coprecipitation system suddenly deteriorated to a severe extent, prompting a re-examination of the problem. It was discovered that the water used for chemical make-up occasionally contained excessive concentrations of sulphate. Consequently, barium sulphate precipitate was being formed in the chemical tank rather than in the coprecipitation reactors and the performance of the coprecipitation operation was adversely affected. The water supply was subsequently changed, resulting in improved performance. Data generated during the upset period were deleted from the computation of means. The mean dissolved radium-226 activities in the effluents of the five coprecipitation reactors, together with the 95% confidence interval, are plotted in Figure 5. The data summarized in that figure are from the entire demonstration phase of approximately 10 months, excluding the anomalous results obtained over a period of 10 days during July, 1980. The graph represents year-round performance and constitutes the principal conclusion with respect to the coprecipitation operation. The system produced a mean dissolved radium-226 activity of 0.19 Bq/L (5 pCi/L).

3.2 Coagulation, Flocculation and Clarification

The clarification process was conceived of as consisting of the following operations: barium-radium coprecipitation in series-connected stirred-tank reactors, the addition of chemical coagulant in a rapid mix unit, gentle agitation in mechanical flocculation units and solid/liquid separation in a clarifier. The objective of the

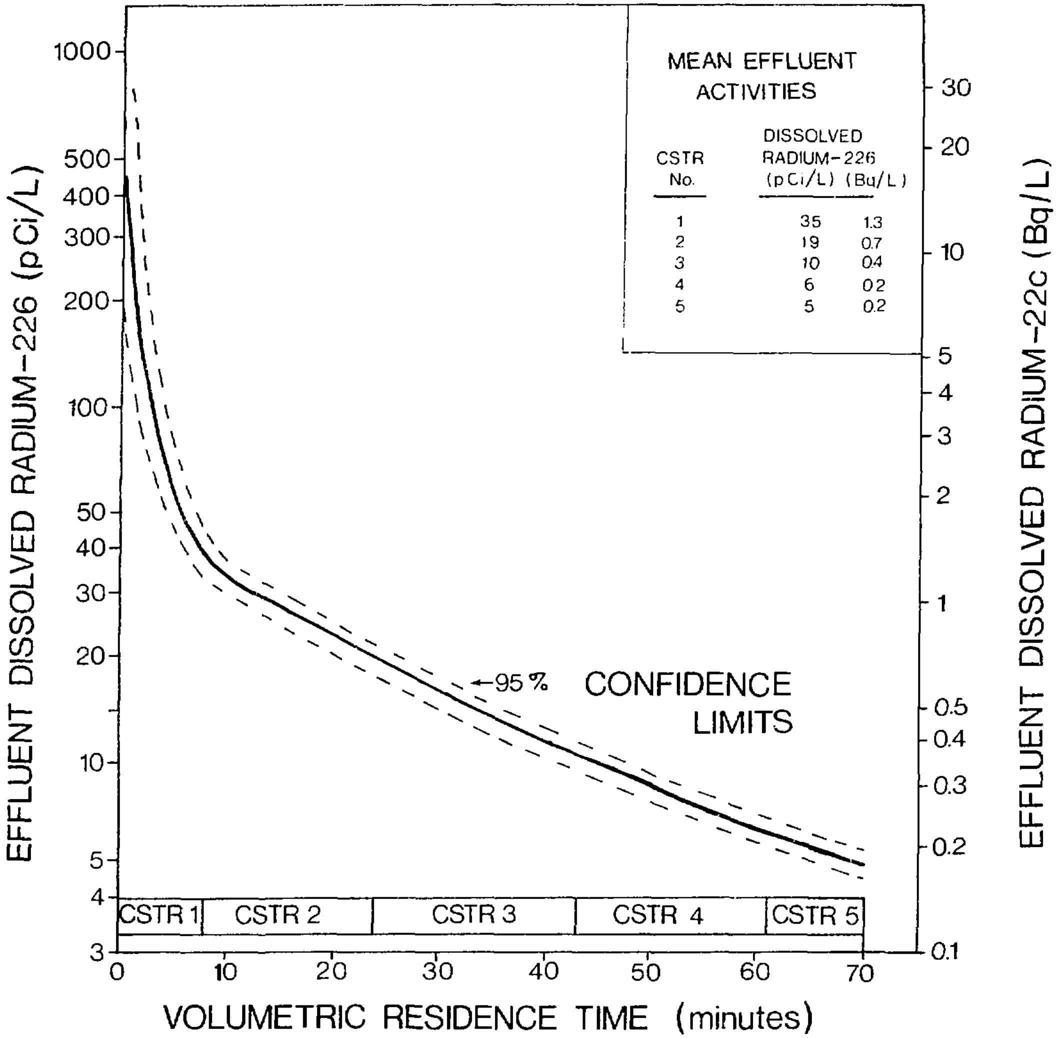


FIGURE 5 COPRECIPITATION SYSTEM PERFORMANCE - DEMONSTRATION PHASE

coagulation, flocculation and clarification operations was the production of a total radium-226 activity less than or equal to 0.37 Bq/L (10 pCi/L) by removal of the (Ba, Ra)SO_4 precipitate formed in the coprecipitation system.

Although the pilot scale development work resulted in a dramatic increase in settling rates, and provided valuable insight into the behaviour of coagulation/flocculation systems, the target of 0.37 Bq/L (10 pCi/L) for total radium-226 activity was not attained consistently. Work on the clarification process was terminated in June, 1980, during the process demonstration phase of the program. The results of the clarification process experiments are not discussed in this report but are presented in Volumes II and III.

3.3 Filtration

Granular media filtration was investigated as an alternative solid/liquid separation operation for achieving a total radium-226 activity 0.37 Bq/L (10 pCi/L) in the process effluent. All filter development work was undertaken at pilot scale using filter columns of at least a 10 cm diameter.

The filtration program examined several design alternatives, including direct filtration versus coagulation/flocculation and filtration, pressure versus gravity operation, single-medium versus dual-media filter beds, and unaided versus chemically-aided filtration. The media sizes were established on the basis of prior experience. The media consisted of silica sand with an effective size of 0.4 mm used either alone or in combination with crushed anthracite coal having an effective size of 1.2 mm. Media depths were established arbitrarily within the normal design range. Ferric chloride and an anionic polymer were tested as filtration aids on the basis of jar test results.

The results of initial process development experiments indicated that direct filtration of coprecipitation system effluent was feasible using a chemically-aided granular media gravity filter. A filtration rate of 6.5 $L/m^2{\cdot}s$ was possible using a dual-media filter bed. Subsequently, process development experiments were conducted over approximately five months to identify possible design and operational problems which would not have become apparent during the initial short-term tests. Uncertainties were associated with the requirement for polymer conditioning of the filter bed, the scale forming tendency of the wastewater (particularly gypsum scale) and the efficiency of the backwash operation. A 20 cm diameter filter (Figure 6) was operated continuously for approximately 165 days; four 24-hour filter runs were monitored per week. The operating conditions for this development period are summarized in Table 4. Although the performance of the filter was initially very good, a gradual increase in both the initial and

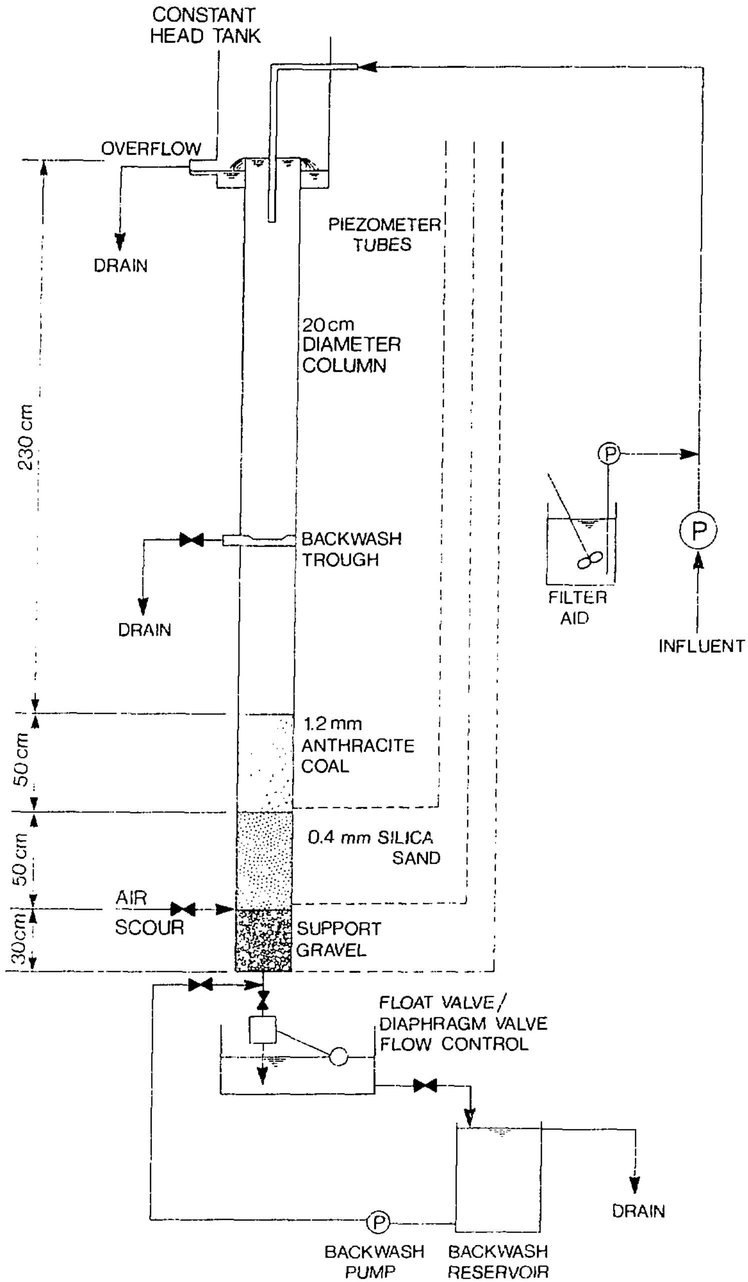


FIGURE 6 DUAL-MEDIA FILTER

TABLE 4 DUAL-MEDIA FILTRATION OPERATING CONDITIONS

	Development Run No.						Demonstration Run No.							
							Period 1					Period 2		
Operating Conditions	10	20	30	40	50	60	70	80	90	100	110	120	130	140
Apparatus & Operation:														
• 20 cm diameter gravity filter • constant head (total available head =3.6 m) • 6.5 $L/m^2 \cdot s$ filtration rate (constant) • 24 hour run length	→	→	→	→	→	→	→	→	→	→	→	→	→	→
Filter bed:														
• coal, 30 cm depth (e.s. = 1.2 mm)*	→	→	→											
45 cm depth (e.s. = 1.2 mm)				→										
50 cm depth (e.s. = 1.2 mm)					→	→	→	→	→	→	→	→	→	→
• sand, 60 cm depth (e.s. = 0.4 mm)	→	→	→	→										
50 cm depth (e.s. = 0.4 mm)					→	→	→	→	→	→	→	→	→	→
• support, 15 cm depth (uniform)	→	→	→	→										
30 cm depth (graded)					→	→	→	→	→	→	→	→	→	→
Monitoring:														
• *headloss; filtration rate; influent SS & dissolved Ra-226;* effluent SS, dissolved Ra-226 & total Ra-226	→	→	→	→	→	→	→	→	→	→	→	→	→	→
• bed height							→	→	→	→	→	→	→	→
Polymer (Percol 727):														
• conditioning of bed by soaking with 1 mg/L polymer solution for 1 hour	→	→	→	→	→	→								
• *dosage = 0.01 mg/L for 1 hour every 8 hours*	→	→	→	→	→	→								
= 0.01 mg/L continuously (except dosage optimization test)							→	→	→	→	→	→	→	→
Backwash:														
• 1/3 bed expansion (19 $L/m^2 \cdot s$) maintained until bed appeared to be clean	→	→	→	→	→	→	→	→	→	→	→	→	→	→
• assisted bed settling (*slow termination* of backwash with vibration of column)	→	→	→	→	→									
• unaided bed settling (sudden termination of backwash with no vibration)						→	→	→	→	→	→	→	→	→
• air scour = 1.2 $m^3/min \cdot m^2$ (moderate)	→	→	→	→	→	→	→	→	→	→	→			
= 6.1 $m^3/min \cdot m^2$ (aggressive)												→	→	→

* e.s. = effective size (diameter of particle at which 10% of particles are smaller)

terminal headloss across the filter was observed over several weeks. The increased headloss was caused in part by mechanical problems in the underdrain system, although a significant operational problem was also identified. That problem appeared as media expansion and intermixing which could be associated with either increasing headlosses or poor effluent quality depending upon the amount of effort exerted to settle the bed after backwashing. Factors contributing to the problem were thought to include polymer accumulation and chemical scale formation on the media particles. Acid washing, with agitation provided by an air scour system, was tested and was believed to be a feasible method of cleaning the filter. However, the frequency and intensity of such cleaning operations remained to be determined.

The goals for the process demonstration phase included long-term demonstration of acceptable effluent quality and identification of process reliability and media cleaning requirements. Demonstration was begun in late February, 1980 and continued to the end of July, 1980. The demonstration phase consisted of two operating periods, each beginning with an attempt to select the optimum continuous polymer dosage. Both operating periods were initiated with fresh anthracite and sand media. The results for the demonstration phase are illustrated in Figure 7; only monitored filter runs are shown.

The operating conditions chosen for the first demonstration period are summarized in Table 4. It was initially determined that acceptable performance could be achieved using a continuous 0.01 mg/L dosage of anionic polymer applied to the influent stream without prior polymer conditioning of the filter bed. Excellent effluent quality was obtained during the next three months. The average total radium-226 activity in the filter effluent stream was 0.11 Bq/L (3 pCi/L) and the average suspended solids concentration was 0.4 mg/L (run numbers 66 to 113). Dissolved radium-226 was also removed by the filter. The mean dissolved radium-226 activity of 0.19 Bq/L (5 pCi/L) from the coprecipitation system was reduced to 0.07 Bq/L (2 pCi/L) in the filter effluent. However, as illustrated in Figure 7, the terminal headloss exceeded its chosen maximum value of 250 cm for Runs 89 through 91. A gradual increase in media expansion over this period was also evident. Thus, although the filter was producing an effluent of acceptable quality, the poor performance characteristics tentatively attributed to polymer accumulation and chemical scale formation were being observed. The filter bed was washed with sodium chloride prior to Run 92 in an attempt to remove accumulated polymer. Headloss observations indicated that the salt and/or the accompanying air scour agitation had some beneficial effect. The results of this demonstration period also suggested that the acid wash frequency would not be greater than once in three months.

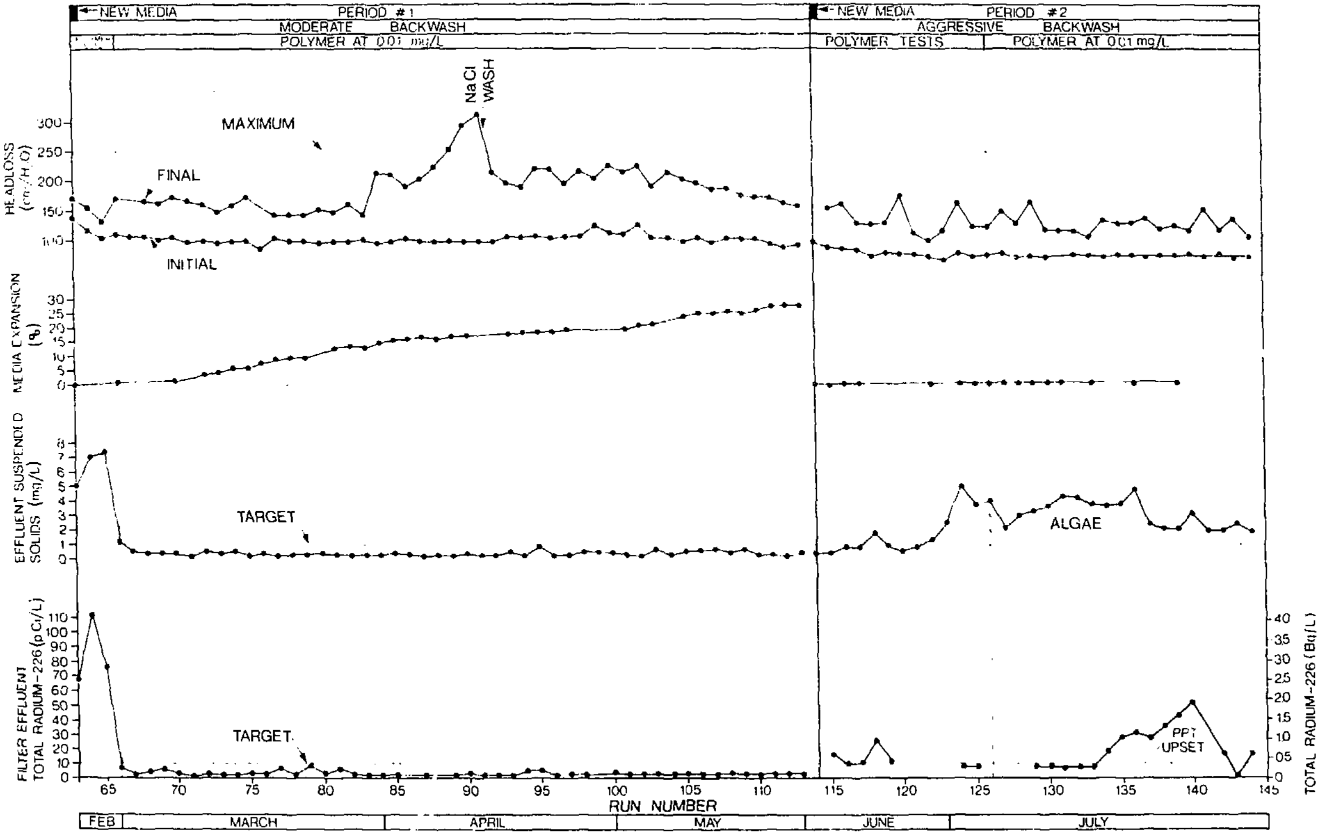


FIGURE 7 DUAL-MEDIA FILTER DEMONSTRATION RESULTS

After Run 113, the filter was taken out of service and samples of the anthracite and sand were removed for laboratory examination. The results showed an accumulation of calcium and barium salts. Batch tests demonstrated that washing the media with a HCl or HNO_3 solution was effective in removing most of this scale.

Fresh media were added to the filter column and the demonstration phase resumed with Run 114. Operating conditions were the same as during the previous runs except that a more aggressive backwashing procedure was initiated with the expectation that polymer accumulation and/or scale buildup on the media grains would be minimized. Operating details are given in Table 4. The second operating period suffered from two problems which did not originate with the filtration operation. An algae bloom in the tailings basin resulted in suspended solids which were not removed by the granular media filter. The consequent change in the solids-to-radium ratio in the filter effluent caused some confusion with respect to the operation of the pilot plant, particularly because of the long lag times involved in obtaining radium-226 results and the necessity of making short-term operating decisions on the basis of suspended solids analyses alone. Secondly, the problem of chemical make-up water quality in the coprecipitation system was reflected in elevated total radium-226 activities measured in the filter effluent. These factors resulted in a lack of meaningful effluent data during this period. However, the headloss and bed height observations indicated clearly that the modified backwash procedure had overcome the problem of filter bed instability. At the end of the demonstration period there was no significant intermixing of the sand and anthracite media, although microscopic examination revealed that scale formation on the media had not been entirely eliminated.

A series of process sensitivity tests followed the demonstration phase. An extended filter run at 6.5 $L/m^2 \cdot s$ was conducted under normal operating conditions to provide a baseline for comparison (Figure 8). The filtration rate and the influent suspended solids concentration were varied in other filter runs. A maximum filtration rate of 9.8 $L/m^2 \cdot s$ was determined to be feasible. Furthermore, the filter demonstrated little sensitivity to varying loading conditions and tended to fail by plugging rather than by solids (or radium) breakthrough.

3.4 Sludge Characterization and Dewatering

The sludge from the clarification process consisted primarily of barium sulphate, calcium carbonate, and ferric hydroxide. The sludge from the filtration process consisted primarily of barium sulphate. The specific activities of the clarification and

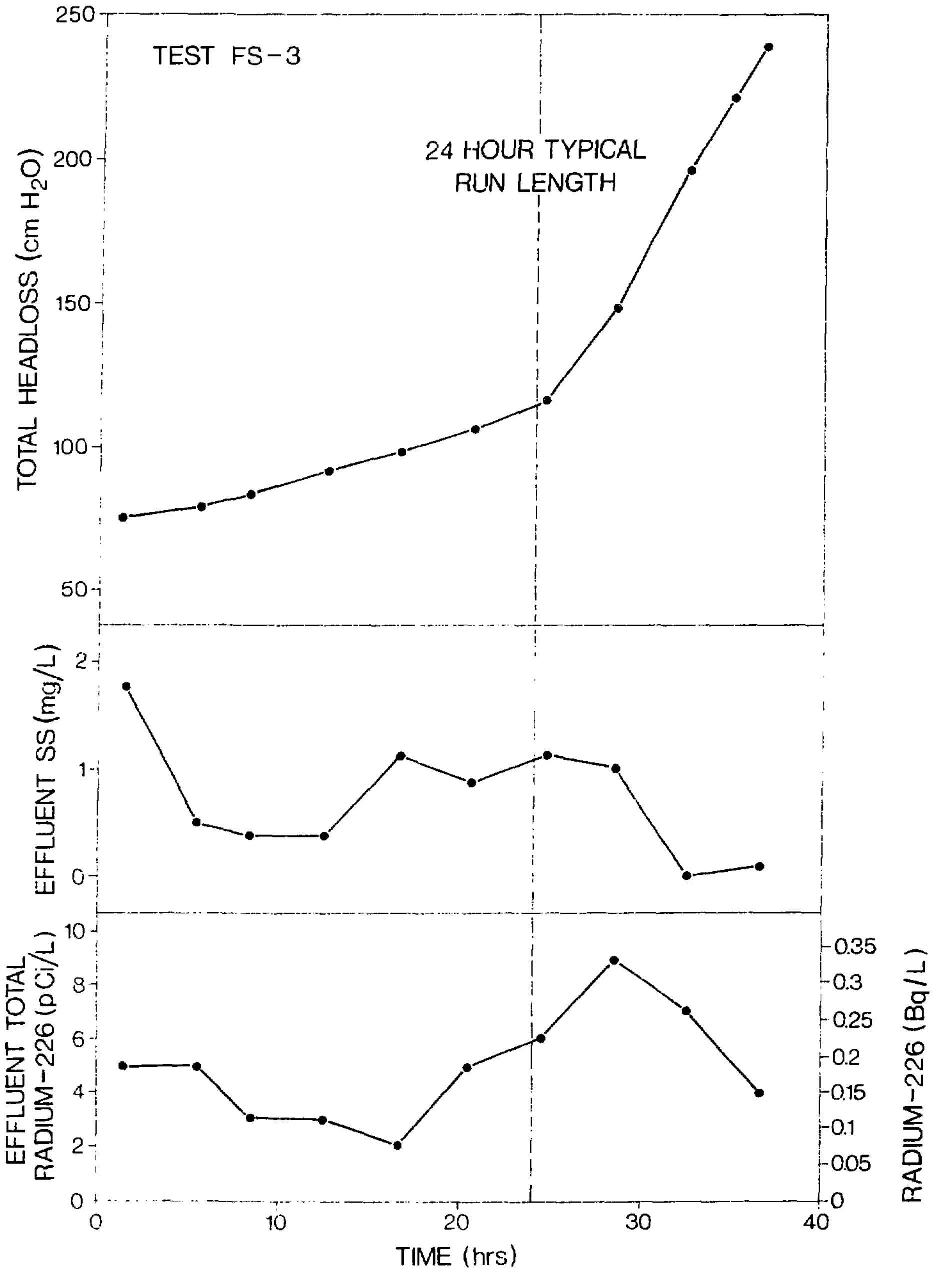


FIGURE 8 TYPICAL DUAL-MEDIA FILTER PERFORMANCE

filtration process sludges were 377 Bq/g (10 200 pCi/g) and 1 017 Bq/g (27 500 pCi/g), respectively (dry weight basis).

The sludge obtained from the clarification process dewatered readily. The greatest volume reduction was effected by the freeze-thaw process although both vacuum filtration and pressure filtration appeared to be potential dewatering methods. The backwash sludge from the filtration process was much more readily dewatered; a 45% solids concentration could be attained by gravity sedimentation.

4 DISCUSSION

4.1 Barium-Radium Coprecipitation

The experimental procedures, which included the principles of similitude and response surface optimization methodology, resulted in close agreement between the laboratory and pilot plant results. Both the bench scale continuous-flow apparatus and the pilot scale CSTR system produced a mean dissolved radium-226 activity of 0.19 Bq/L (5 pCi/L).

The dissolved radium-226 activity was significantly reduced as the wastewater stream proceeded through each of five series-connected reactors. Although statistically significant over a period of approximately ten months, a difference of only 0.04 Bq/L (1 pCi/L) between the fourth and fifth reactors was not considered to be sufficiently large to warrant a fifth reactor. Therefore, the following reactor system was selected: four equal-size CSTR's in series each having a volumetric residence time of 15 minutes. The performance of this four-reactor system, as well as alternative three-reactor and five-reactor systems, were predicted using reaction rate data obtained from the pilot plant. The alternative reactor systems are expected to produce a dissolved radium-226 activity of approximately 0.22 Bq/L. This level of performance would be anticipated at any scale, providing that geometric, dynamic and chemical similarly with the pilot plant system are maintained.

4.2 Filtration

The filtration process was considered to have been satisfactorily demonstrated during the two operating periods in the demonstration phase. During the first period, the filter produced an effluent of very good quality at a filtration rate which is generally considered to be well within the range of technical and economical feasibility. The problem of filter bed instability (expansion and intermixing) experienced during the first operating period was subsequently overcome by a more aggressive backwash operation. Unfortunately, two factors external to the filtration operation, algae in the plant influent and an upset in the coprecipitation system, limited the amount of meaningful effluent data available from the second operating period.

Full-scale filtration systems are commonly designed on the basis of pilot scale studies using filter columns of at least a 10 cm diameter. Pilot scale models of that size reasonably represent the performance of the filter in filtration mode, although the models are not adequate to represent backwash performance (USEPA, 1975). Therefore, the filtration performance experienced at pilot scale is anticipated to occur at full scale. However, since filter cleaning was identified as a potential problem and the pilot plant was too small for adequate backwash modelling, the specifications for full-scale backwashing and acid cleaning operations should include a considerable amount of flexibility.

The data base for the design of a full-scale process is presented in Table 5. Figure 9 is a schematic representation of the process.

TABLE 5 DATA BASE FOR PROCESS DESIGN

Barium-Radium Coprecipitation	Barium dosage*	16 mg/L as Ba
	Precipitation reactors: (alternative systems) No. of CSTR units and volumetric residence time	 3 @ 26.7 = 80 min 4 @ 15.0 = 60 min 5 @ 11.0 = 55 min
	Precipitation power input	6×10^{-2} kW/m^3
Granular Media Filtration	Filter aid dosage**	0.01 mg/L
	Filtration rate: design maximum	$6.5\ L/m^2 \cdot s$ $9.8\ L/m^2 \cdot s$
	Filter bed depth: anthracite sand	0.5 m 0.5 m
	Anthracite specifications: - effective size - uniformity coefficient - maximum size	 1.2 mm 1.6 4.8 mm
	Silica sand specifications: - effective size - uniformity coefficient - maximum size	 0.4 mm 1.6 1.2 mm

* Barium must be in an available, dissolved form. Sulphate in the chemical make-up water will precipitate barium and reduce process efficiency.

** Continuous flow of high molecular weight anionic polymer; optimum polymer type and dosage may be site-specific.

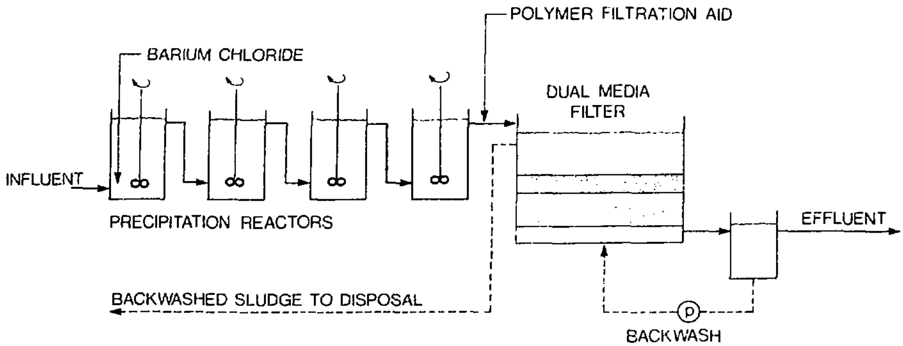


FIGURE 9 RADIUM-226 REMOVAL PROCESS

5 COST ESTIMATION

Capital costs and operating and maintenance costs were estimated for three full-scale barium-radium coprecipitation and filtration plants. The plant sizes selected were 380, 150 and 26 L/s.

To initiate the cost estimation procedure, a cost estimation data base was derived from Table 5 by the inclusion of all ancillary equipment required to make a treatment plant functional. The cost estimation data base and supplementary design notes are included in Volumes II and III. Process and instrumentation diagrams were prepared in conjunction with the cost estimation data base (Figures 10 and 11). Equipment layout sketches were drawn for each of the three plant sizes; Figure 12 is the general layout of the largest treatment plant. Two sludge handling options were considered:

- Option 1 includes batch thickening of backwash sludge as preparation for some undefined ultimate disposal operation.
- Option 2 includes provision for transporting the backwashed sludge in slurry form to the tailings basin.

5.1 Capital Cost Estimates

The capital cost estimates were prepared by consulting engineers with experience in the design and construction of wastewater treatment plants (MacLaren, 1981). Equipment costs were obtained from suppliers and were based on delivery in March 1981. Site preparation and building costs were prepared on the basis of experience in the Elliot Lake area.

The capital cost estimates for the 380 L/s plant size are summarized in Table 6.

5.2 Operating and Maintenance Cost Estimates

The operating and maintenance (O&M) cost estimates were prepared at the WTC. Labour requirements and maintenance material costs were based on published data for water and wastewater treatment plants (Gumerman et al., 1978; Patterson and Banker, 1971). Published costs were updated to the spring of 1981. A total payroll cost of \$20/hour was assumed to apply to O&M labour. The following unit costs were employed: power at 3.1 cents per kilowatt-hour, barium chloride at \$20 per 50 kg bag, polymer at \$5 per kg, 60% acid (HCl or HNO_3) at \$77 per cubic metre. See Volume III, Appendix V for further details.

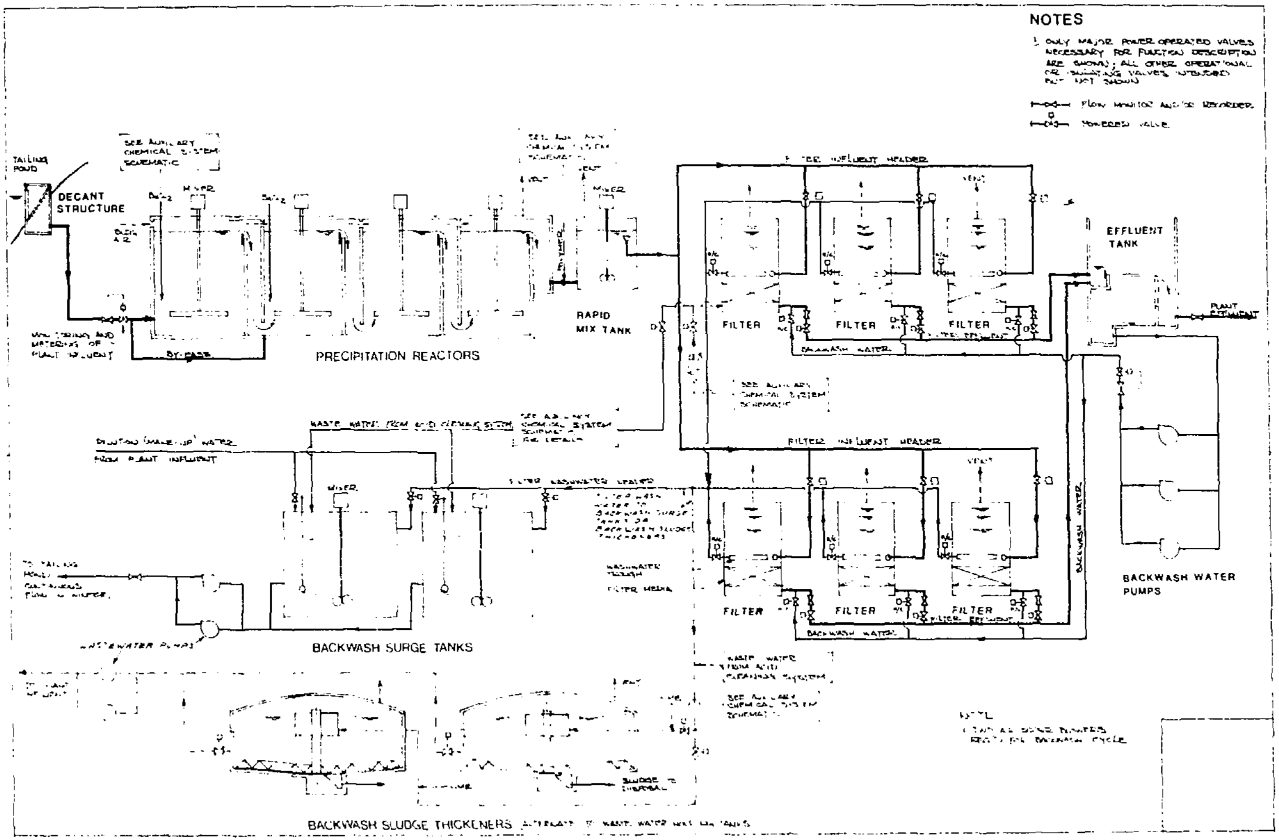


FIGURE 10 SIMPLIFIED PROCESS FLOW SCHEMATIC

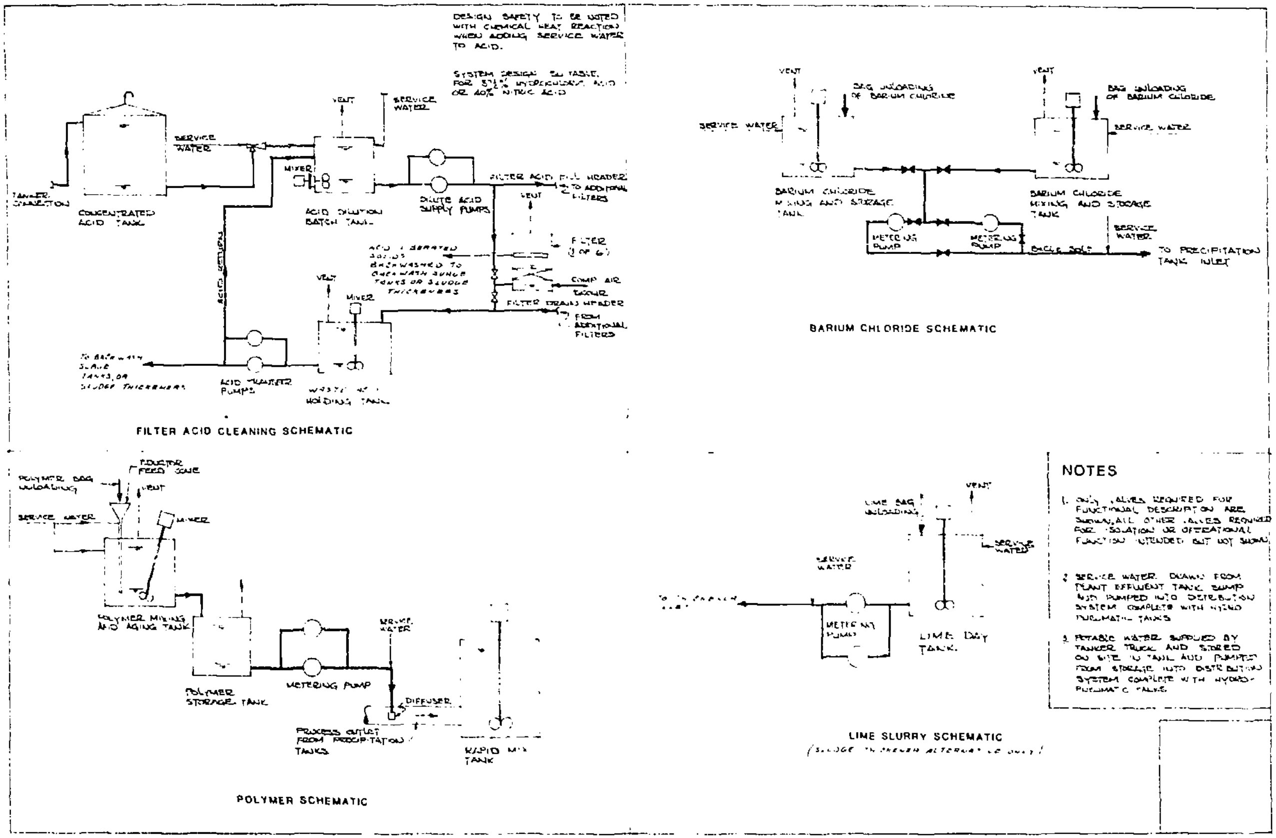
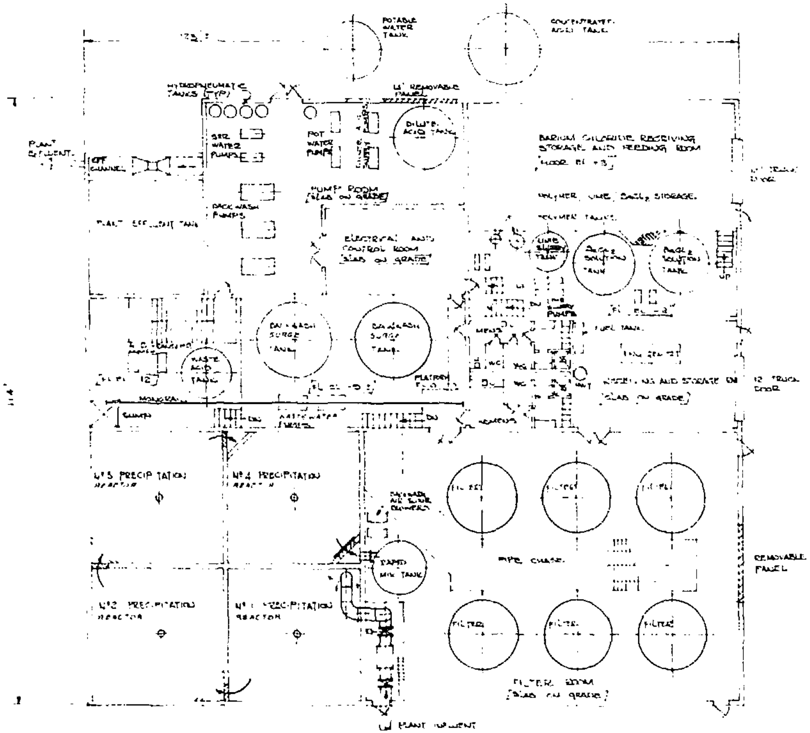


FIGURE 11 SIMPLIFIED AUXILIARY CHEMICAL SYSTEM SCHEMATIC



380 L/S PLANT LAYOUT

NOTES

1. THIS TYPICAL PLANT LAYOUT IS BASED ON THE DESIGN OF A 380 L/S PROCESS. SIMILAR FEATURES WOULD BE REQUIRED IN THE SMALLER PLANTS.

2. A PREFABRICATED METAL STRUCTURE IS ASSUMED TO ENCLOSE ALL FACILITIES. THE PRECIPITATION TANKS, POTABLE WATER TANK AND THE CONCENTRATED ACID TANK WOULD BE LOCATED OUTSIDE.

FIGURE 12 TYPICAL PLANT LAYOUT

TABLE 6 CAPITAL COST ESTIMATE SUMMARY - 380 L/s PLANT

Item	Detail	Cost (1981 Dollars) Option I	Option II
Building	Structural & Architectural	622 000	622 000
Building Services	Plumbing	66 650	66 650
	Heating, Ventilation	108 200	108 200
	Lighting	83 000	83 000
Mechanical Process	Precipitation Reactors	445 116	445 116
	Acid Mix Tank	17 025	17 025
	Filters (6)	406 200	406 200
	Effluent Storage	13 025	13 025
	Sludge Thickeners	70 550	
	Backwash Surge Tanks		47 110
	Barium Chloride System	51 490	51 490
	Polymer System	20 120	20 120
	Lime System	11 295	
	Acid System	44 655	44 655
	Process Pumps	12 375	12 375
	Process Mixers	8 100	8 100
	Piping and Valves	449 200	449 200
Miscellaneous Mechanical Equipment	Loader, Generator Set	108 000	108 000
Electrical	Sub Station	70 000	70 000
	Power Wiring	321 550	321 550
Instrumentation and Wiring	Components, Boards, Etc.	286 800	286 800
Sub Total		3 215 351	3 180 616
Contingency	20%	643 070	636 123
Sub Total		3 858 421	3 816 739
Engineering	10%	385 842	381 674
TOTAL COST		4 244 263	4 198 841

The operating and maintenance cost estimates for the 380 L/s plant size are summarized in Table 7.

TABLE 7 OPERATING AND MAINTENANCE COST ESTIMATE SUMMARY - 380 L/s PLANT

Item	O&M Cost ($1 000's/year) (Spring, 1981)
PRECIPITATION	
- power	18.2
- $BaCl_2$	136.0
FILTRATION	
- power	1.8
- parts	5.4
- polymer	0.6
- acid	6.1
- labour	110.0
SLUDGE	
- pumping	3.5
- labour	28.0
- thickeners or surge tank	11.0
TOTAL	320.6

5.3 Total Cost Estimates

Total annual costs were estimated based on the Option 1 capital and O&M costs (spring, 1981) using a 15-year amortization period and a 15% interest rate calculated yearly:

- 380 L/s Plant $1 046 400
- 150 L/s Plant $ 710 200
- 26 L/s Plant $ 401 400

6 CONCLUSIONS AND RECOMMENDATIONS

6.1 Conclusions

With reference to Table 2, goals 1, 2 and 3 were achieved. A physical/chemical wastewater treatment process was developed, and demonstrated at pilot scale to produce an effluent meeting the program targets of 0.37 Bq/L (10 pCi/L) for total radium-226 and 0.11 Bq/L (3 pCi/L) for dissolved radium-226. Principal design specifications are provided in Table 5. Goals 4 and 5 were partially achieved; sludge characterization and dewatering experiments were undertaken in a limited bench scale program. Pilot scale sludge dewatering tests were considered to be beyond the scope of the program in view of pending decisions by others with respect to suitable sludge disposal options. Goal 6 was partially achieved in that capital and operating/maintenance costs were estimated for the wastewater treatment process using a range of plant sizes.

The principal factors in the barium-radium coprecipitation operation were the barium dosage and the reactor residence time. Attainment of the program target of 0.11 Bq/L (3 pCi/L) on a continuous basis was not feasible within the coprecipitation reactor system. The reactor system demonstrated at pilot scale produced a mean dissolved radium-226 activity of 0.19 Bq/L (5 pCi/L). The recommended alternative reactor configurations are expected to produce a dissolved radium-226 activity of approximately 0.22 Bq/L (6 pCi/L).

Solid/liquid separation was achieved by granular media filtration of the coprecipitation system effluent. A gravity, dual-media filter was operated at a filtration rate of 6.5 $L/m^2 \cdot s$ using a high molecular weight anionic polymer as a filter aid. The filter produced an effluent with a total radium-226 activity of less than the target of 0.37 Bq/L (10 pCi/L) and a dissolved radium-226 activity of less than the target of 0.11 Bq/L (3 pCi/L). Filter runs were terminated by plugging of the bed, rather than by solids or radium breakthrough; thus, it was possible to maintain consistent effluent quality.

Filter cleaning operations were found to be a very important design consideration, although the size of the pilot scale equipment did not permit precise quantification of the cleaning operations. Backwashing, with supplementary cleaning by air scour, was required approximately once every twenty-four hours under normal operating conditions. An aggressive backwash operation was shown to overcome the problem of filter bed instability (media intermixing and bed expansion) but did not eliminate scale accumulation on the media or other surfaces of the filter apparatus. Periodic acid washing with 2% (V/V) hydrochloric or nitric acid was required to remove acumulations of scale. The acid cleaning frequency was not expected to exceed four operations per year.

The backwash sludge from the filtration process consisted primarily of barium sulphate and had a specific activity of approximately 1 017 Bq (27 500 pCi) per gram of dry solids. Bench scale tests indicated that the sludge was readily dewaterable.

6.2 Recommendations

A full-scale prototype plant incorporating coprecipitation and filtration operations has been built at the Stanleigh Uranium Mine in Elliot Lake, Ontario by Rio Algom Limited. It is recommended that extensive monitoring be undertaken during the first two or three years of operation.

Additional experimental work is required to obtain a better understanding of the physical and chemical mechanisms involved in the process and to assess other potential process alternatives. Although important, this work was considered to be beyond the scope of this program and is recommended for further study.

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MEMBERS OF THE MANAGEMENT PANEL AND TECHNICAL WORKING GROUP

Management Panel:

Mr. J. Schmidt	Environment Canada (Chairman)
Mr. E. Barnes	Rio Algom Limited
Mr. E. Larocque	Denison Mines Limited
Dr. A. Ashbrook	Eldorado Resources Limited[1]
Dr. J. Coady	Atomic Energy Control Board
Mr. J. Scott	Environment Canada
Mr. W. Craigen[2]	Energy, Mines & Resources Canada
Mr. P. Kresin	Madawaska Mines Limited
Mr. A. Morrish	Gulf Minerals Canada Limited
Mr. P. Fournier	Amok Limited
Mr. M. Neven[3]	Key Lake Mining Corporation

Technical Working Group:

Mr. D. Averill	Environment Canada (Chairman)
Dr. D. Moffett[4]	Rio Algom Limited
Mr. R. Webber	Denison Mines Limited
Mr. R. McClure[5]	Eldorado Resources Limited

1 Formerly Eldorado Nuclear Limited.
2 Formerly represented by Dr. G. Herget.
3 Formerly represented by B. Rosner, Uranerz Exploration and Mining Limited.
4 Currently with Eldorado Resources Limited.
5 Formerly represented by Dr. V. Lakshmanan (1978) and Dr. G. Kassakhian (1979).

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