

BRIEF PRESENTED
TO
THE BRITISH COLUMBIA
ROYAL COMMISSION OF INQUIRY
HEALTH AND ENVIRONMENTAL PROTECTION
URANIUM MINING

PHASE V

by

Department of Environment
Department of Fisheries and Oceans

Environmental Protection Service
Department of Environment
Pacific Region

Report EPS 7-PR-79-3

November, 1979

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URANIUM MINING INDUSTRY - POLLUTION CONTROL RESEARCH

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URANIUM MINING INDUSTRY - POLLUTION CONTROL RESEARCH

1 INTRODUCTION

Environment Canada is currently carrying out two major research projects at its Wastewater Technology Centre (WTC) in Burlington, Ontario. These projects deal with: 1) the physical- chemical removal of Ra-226 from mill effluents (Section 2) and; 2) the leachability of radioactive constituents from uranium mine tailings (Section 3).

In addition, Section 4, offers brief description of a number of other research projects being carried out in Canada. The listing is restrictive in the sense that it pertains to research relating to potential technologies or measures to reduce the release of contaminants generated during the mining and milling of uranium. It does not address, for example, the research being undertaken on environmental impacts of contaminants, environmental pathways, etc.

2 PHYSICAL-CHEMICAL REMOVAL OF RADIUM-226 FROM MILL EFFLUENTS

The Wastewater Technology Centre initiated a study in 1975 to develop improved methods of removing radionuclides from effluents from uranium mines and mills. The study was designed to refine and upgrade existing technology rather than to replace current methods with completely new approaches.

In March 1976, preliminary bench scale experiments were initiated to better understand and improve the Ra-226 removal process. During 1977, the Atomic Energy Control Board, encouraged by the initial success of the study, assisted in accelerating the program by providing additional funds.

Based on these preliminary studies, a jointly managed and funded government/industry project was developed. The project is financially supported by Rio Algom Ltd., Madawaska Mines Ltd., Denison Mines Ltd., Eldorado Nuclear Ltd., AMOK Ltd., the AECB, Environment Canada and the Federal Department of Energy, Mines and Resources. Gulf Minerals Canada Ltd. and Key Lake Mining Corporation are also participants. The specific goals developed for the program are as follows:

1. To develop at pilot scale, a physical-chemical process to reduce the Ra-226 content of uranium mining and milling effluents.
2. To demonstrate at pilot scale, a reasonably achievable level of Ra-226 in the effluent, with target levels of 10pCi/l total Ra-226 and 3pCi/l dissolved Ra-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.

The project included both bench scale and pilot scale experimental programs. Two treatment process alternatives were examined; the first consisted of barium-radium coprecipitation followed by coagulation, flocculation and sedimentation (Figure 1). The second process consisted of barium-radium coprecipitation followed by granular media filtration (Figure 2).

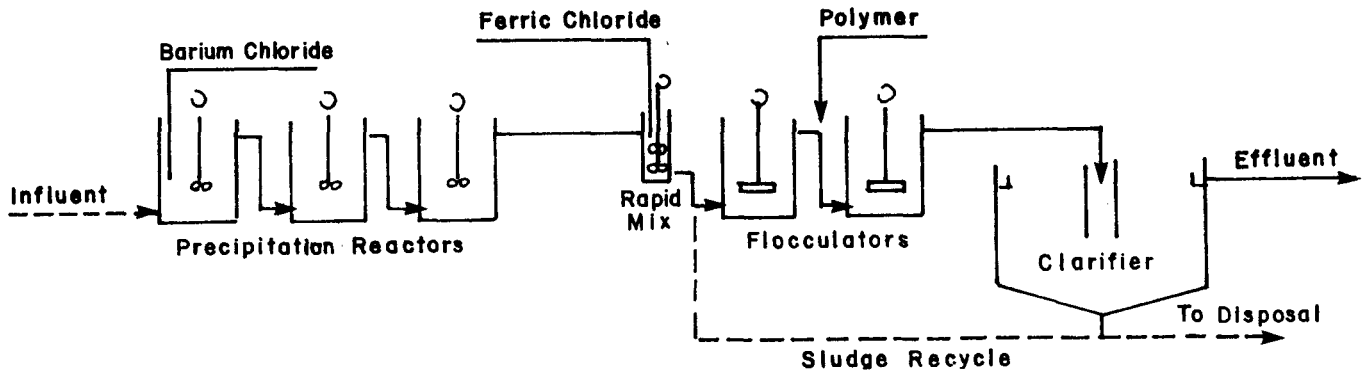


FIGURE 1 SCHEMATIC OF CLARIFICATION PROCESS

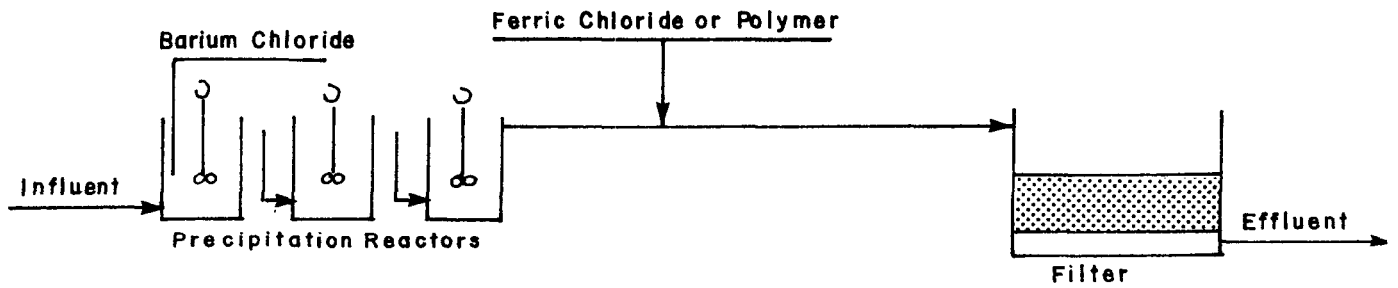


FIGURE 2 SCHEMATIC OF FILTRATION PROCESS

2.1 Bench Scale Program

The bench scale experiments were conducted at the Wastewater Technology Centre in Burlington, Ontario. The purpose of these experiments was to identify optimum operating conditions for the coprecipitation of radium with barium sulphate and for chemical coagulation and flocculation of the resulting BaRaSO₄ suspension. Both batch and continuous flow techniques were employed. Samples of a tailings area decant were obtained from an acid leach uranium milling operation. Ra-226 activity level determinations were performed by radon de-emanation.

For the precipitation operation, batch experiments were conducted to identify the process variables having a significant effect on soluble radium activity levels in the effluent and to determine optimum operating conditions. Kinetic experiments were performed to determine the order and rate of the precipitation reaction. The information obtained was used to size reactors for continuous flow experiments which were conducted to confirm the results of the batch experiments. The optimum precipitation conditions selected for further testing at pilot scale consisted of barium chloride dose of 16 mg/l (as Ba) and a retention time of 110 to 120 minutes in three series-connected stirred tank reactors.

Similar experiments were undertaken to develop the coagulation/flocculation operation. Ferric chloride was chosen as the coagulant at a dosage of 4 mg/l (as Fe). There was considerable flexibility in the rapid mix time; and the optimum contact time, in two continuous flow series-connected mechanical flocculation units, was determined to be 80 minutes. Sludge recycle from the clarifier underflow to the influent of the first flocculator was found to be beneficial when employed in conjunction with a polymeric flocculation aid.

2.2 Pilot Scale Program

Pilot scale experiments were undertaken at the Rio Algom Ltd., Quirke Mine to investigate both the clarification and filtration solids separation alternatives. Mobile physical-chemical wastewater treatment facilities with a nominal capacity of 5 gpm were used. With respect to the clarification process, the objectives of this series of experiments were: 1) to verify the results of the bench scale tests, 2) to identify scale-up relationships and 3) to provide information on operating conditions not readily available from bench scale systems. Results of the development phase of the clarification process studies are presented in Tables 1 and 2. Development of the filtration process was undertaken exclusively at pilot scale.

In Table 1 the data on a "once-through" process involving precipitation, coagulation and solid/liquid separation using a conventional circular clarifier (Experiment 1) is presented (1). Data are also presented on a similar process involving sludge recycle from the clarifier to the flocculators and also the addition of a polymer (1). In Table 2 the results of a process involving precipitation, coagulation, polymer addition, and solid/liquid separation using a "plate" clarifier with sludge recycle from the clarifier to the flocculators are presented (1). The target dissolved Ra-226 level of 3 pCi/l was met when the hydraulic loading ranged from 600 to 2000 lgal/ft²/d and the flocculator suspended solids (design) concentration was 3000 mg/l.

With regard to the development of the filtration process, several screening experiments have been conducted using pressure and gravity operational modes, single and dual media beds, several hydraulic loading rates and two chemical filtration aids at several dosages. At this time, it appears that effluent total Ra-226

TABLE 1 CLARIFICATION PROCESS PERFORMANCE - CONVENTIONAL CIRCULAR CLARIFIER*

	Experiment 1	Experiment 3		
	Once-Through Treatment Mode (Baseline)	Sludge Recycle Only	Recycle + 0.4 mg/l Polymer	Recycle + 1.2 mg/l Polymer
Influent Total Ra-226 (pCi/l)	1080 (148)	783 (81)	783 (70)	999 (192)
Precipitation Effluent Dissolved Ra-226 (pCi/l)**	11 (5)	6 (1)	5 (3)	3 (1)
Clarifier Effluent Total Ra-226 (pCi/l)	49 (20)	32 (11)	22 (5)	18 (3)
Clarifier Effluent Dissolved Ra-226 (pCi/l)**	6 (3)	7 (4)	4 (3)	3 (1)

* Results expressed as mean and (standard deviation).

** Dissolved - 0.45 micron filter.

TABLE 2 TOTAL Ra-226 ACTIVITY IN THE PROCESS EFFLUENT - PLATE CLARIFIER WITH SLUDGE RECYCLE*

Flocculator Suspended Solids	Hydraulic Load (Gal/ft ² /d)							
	600	900	1200	2000	3500	5000	7500	10000
1500 mg/l***	14 (5)							
3000 mg/l***	8 (3)	9 (2)	7** (1)	5 (2)	8 (3)	8 (1)	8 (2)	7 (2)

* Data presented as mean and (standard deviation) in pCi/l.

** The suspended solids concentration in the flocculators was approximately 2000 mg/l because of operational problems.

*** Approximate concentrations.

activity levels of less than 10 pCi/l can be achieved using granular media filtration at hydraulic loads of up to 7 gpm/ft² (2,3).

2.3 Project Continuation

Development and demonstration of the clarification and filtration processes were continued until September, 1979. Other process options, including sludge recycle in the precipitation operation and the use of floc weighting agents in the flocculation operations, will be investigated at bench scales and tested at pilot scale if warranted. Sludge characterization and dewatering experiments are also planned. Preliminary cost estimates for the various process options are expected to be completed by October, 1979.

3 LEACHABILITY OF RADIOACTIVE CONSTITUENTS FROM URANIUM MINE TAILINGS

In response to technology gaps identified during the development of the Federal Metal Mining Liquid Effluent Regulations and Guidelines a project was initiated by the Wastewater Technology Centre to assess:

- 1) the extent of the leaching of potentially environmentally hazardous radionuclides (Ra-226, Pb-210, Th-228, Th-230 and Th-232) from uranium mine tailings;
- 2) techniques for the ultimate disposal of tailings and related treatment residues such as BaRaSo₄ sediments.

The program was initiated in 1974 and has subsequently been refined and expanded. In Figure 3 an experimental chronology is presented. During the past year three concurrent phases have been actively pursued. A discussion of the work carried out follows (4).

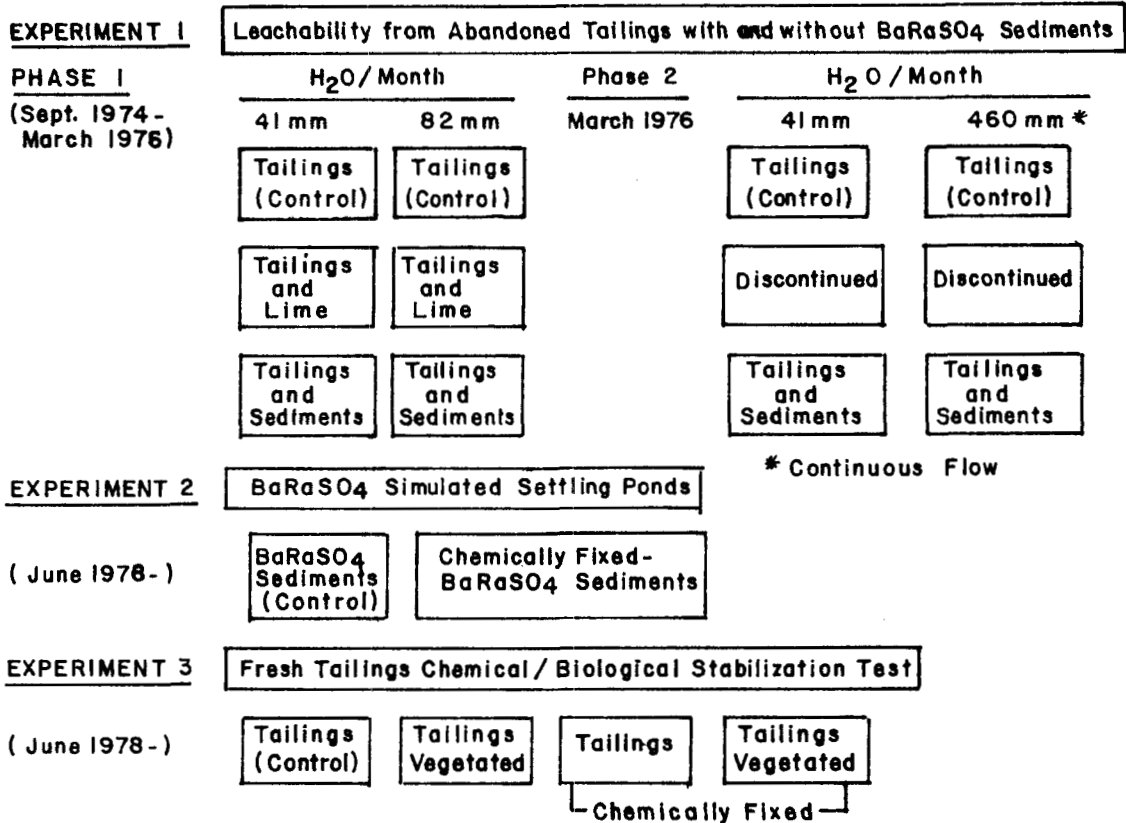


FIGURE 3 RADIOACTIVE LEACHABILITY PROJECT
EXPERIMENT PHASING AND CHRONOLOGY

Experiment 1

The leaching of stored tailings and tailings plus BaRaSO₄ sediments from the treatment of an acidic seepage has been in progress for four years. Two flow rates were used to establish the effect of water flow regime on the rate of release of radioactive constituents. During the past year Ra-226 levels in leachate continued to increase at the high water application rate. This increase in Ra-226 level was greater from the tailings plus BaRaSO₄ sludge than from tailings alone. In September, 1977, the levels averaged 9 and 33 pCi/l compared to levels in June, 1978, of 7 and 76 pCi/l from the low and high water application rates, respectively. Levels of Pb-210 in leachate in September, 1977 averaged 3150 and 940 pCi/l, decreasing by March, 1978, to 2244 and 852 pCi/l for the low and high water application rates, respectively. Levels of thorium in the leachate remain to be determined, and will be reported at a later date. The experiment will continue to run through 1979, but sampling will be reduced to once yearly to establish long term stability.

Experiment 2

Simulated settling ponds have been used to determine the effect of chemical fixation on the release of radionuclides from BaRaSO₄ sediments derived from the treatment of an acidic seepage. This project was initiated in July, 1976. Following 26 months of monitoring, the levels of Ra-226 in effluent have remained stable at about 7 pCi/l for the control compared to 3 pCi/l from the chemically fixed sediments. Levels of Pb-210 have remained 15 pCi/l for all treatments. Bioassay tests conducted on the leachate from the chemically fixed sediments during September, 1978, indicated no toxicity compared to tests during 1977 in which the leachate was toxic to rainbow trout. The reason for the change in the toxicity was not determined.

Experiment 3

A third study was initiated in 1976 in order to establish the rate of radioactive leaching from freshly milled tailings (control) from the Quirke Mine, Elliot Lake, and to reduce this rate by:

1. chemical fixation;
2. sewage sludge application to obtain 3% organic matter in the surface 15 cm followed by revegetation;
3. a combination of 1 and 2.

Levels of Ra-226 in leachate during the first harvest period of 1978 averaged 113 and 27 pCi/l for the non-fixed and chemically fixed tailings, respectively. This represents a decrease from the levels measured in November, 1977. Non-vegetated treatments leached 200 and 149 l/lysimeter compared to 129 and 103 l/lysimeter from vegetated treatments for non-fixed and chemically fixed tailings, respectively. Levels of Pb-210 in leachate during the first harvest in 1978 increased to an average of 384 pCi/l for the non-fixed tailings and remained at 15 pCi/l for the chemically fixed tailings. Levels of thorium in leachate are not yet determined. Yields of grass were similar to low productive soils, producing 2.61 and 3.05 tons/acre from the sludged tailings and chemically fixed sludged tailings respectively. Although the stand of vegetation has remained stable, the decrease in yield (approximately 50% of 1977 yield) indicates a lack of proper nutrient supply. No radiochemical analyses have been performed on the vegetation to date.

3.1 Bench Scale Tests

During 1978, two bench scale tests were also set up. The primary objective of the first bench study was to determine if the solubility of sulfate in tailings directly affects the level of

Ra-226 in the leachate. Various water flow rates, both intermittent and continuous, were applied to tailings from Rio Algom's Quirke Mine and Madawaska's mine. Analysis of the preliminary data is not complete and consequently, cause and effect relationships have not been established. A cursory examination of the data does indicate some generalized trends. The leachate from the Rio Algom tailings dropped from a pH of 8 to less than 4 over a period ranging from 14 to 28 weeks. When the pH dropped below 7, increases of 200 to 300 pCi/l were observed in the radium levels from most treatments. The pH of the Madawaska tailings leachate has remained stable, between 7 and 8, and levels of Ra-226 have continued to decrease. The absolute levels of Ra-226 depend on the water flow rate and the mode of application. Monitoring of the system and data analysis are continuing.

The objective of the second bench scale test was to establish the effect of quiescent and agitated conditions on the solubility rate of Ra-226 from BaRaSO₄ sediments. Due to equipment malfunctions, relocation of the experiment and high evaporation rates, trends could not be identified in the data. It is expected that the experiment will be repeated next year after modifying the operational procedures.

4 AN OVERVIEW OF OTHER URANIUM INDUSTRY POLLUTION CONTROL RESEARCH

In this section, major industry pollution control research projects being carried out in Canada are outlined (5). It is not our intention to describe each project in detail but rather to identify:

- 1) the work being carried out; 2) the organization(s) involved in each project. If additional information on a particular research project is needed, the organization(s) involved in the study should be approached.

Research Category	Research Organization(s)	Brief Description of Research
Mine Water	Eldorado Nuclear Ltd. and Canada Centre for Mineral and Energy Technology, Dept. of Energy, Mines and Resources Canada (CANMET)	<p>1) Joint program on the use of continuous ion exchange for the removal of uranium, chloride and radium. The work is to be carried out at Beaverlodge, Sask. on a pilot scale level.</p> <p>2) Studies are being carried out to determine whether uranium and possibly radium can be precipitated from minewater underground. Bench scale studies have been completed and pilot scale facilities are under construction.</p>
	Rio Algom Ltd. and Denison Mines Ltd.	<p>Commissioned a feasibility study for the removal of nitrogen compounds from mine water. The processes considered for ammonia removal were:</p> <ol style="list-style-type: none"> 1) biological nitrification, 2) breakpoint chlorination, and 3) ion exchange. <p>Nitrate removal by biological denitrification was examined. Results of the study are currently being assessed by the proponents. The mines have also undertaken studies to better quantify nitrogen sources and further examine treatment options.</p>

Research Category	Research Organization(s)	Brief Description of Research
Mill Wastewater (non-radionuclides)	Rio Algom Ltd. and Denison Mines Ltd.	Undertaken research to determine the feasibility of substituting other reagents for those containing nitrogen. Processes examined/ developed include LAMIX, HIMIX and the use of hydrogen peroxide. A report was also commissioned to examine the feasibility of removing nitrogen from the tailings impoundment effluent. A mill process change involving the recovery of sulphuric and nitric acids for re-use was also considered. The mines are currently examining the results of this study.
Mill Wastewaters - Radium Removal (Investigations within the mill)	Rio Algom Ltd., Denison Mines Ltd., Eldorado Nuclear Ltd, and CANMET	Undertaken Ra-226 surveys within the mill to determine sources and potential treatment options.
	Rio Algom Ltd.	Examined the possibility of extracting Ra-226 from the tailings solids prior to discharge to the tailings impoundment facilities.

Research Category	Research Organization(s)	Brief Description of Research
<p>Mill Wastewaters - Radium Removal (Investigations within the mill) (cont'd.)</p>	<p>McMaster University</p>	<p>Examination of a number of extractants to remove radium from the tailings solids prior to discharge to the tailings impoundment facilities. This program has recently received substantial funding from the Natural Science and Engineering Research Council of the Federal Government.</p>
	<p>CANMET</p>	<p>Undertaken research on entirely new uranium extraction processes. For example, a dry chlorination process with a goal of recovering at least 95% of the uranium, 60% of the thorium, 80% of the rare earths and producing barren solids of less than 20 pCi/g Ra-226 has been examined.</p>
<p>Mill Wastewater - Radium Removal (from Tailings Area Effluents)</p>	<p>Eldorado Nuclear Ltd.</p>	<p>Undertaken column settling studies on BaRaS04 precipitates. Also, full scale studies on a number of options have been undertaken at Beaverlodge, Sask. In addition, Eldorado Nuclear Ltd. have recently proposed to undertake basic studies on the precipitation of BaS04 and BaRaS04.</p>

Research Category	Research Organization(s)	Brief Description of Research
Mill Wastewater - Radium Removal (from Tailings Area Effluents) (Contd.)	Rio Algom Ltd.	Undertaken laboratory and pilot scale studies to determine the feasibility of producing an effluent with a Ra-226 activity of less than 10 pCi/l total and 3 pCi/l dissolved. The pilot plant consists of a mixing tank for BaCl ₂ addition, an ageing tank and two ponds (nominal 10 days retention time) for settling of the BaRaSO ₄ precipitate. The nominal flowrate is 50 gpm.
	Madawaska Mines Ltd.	A study was undertaken on the feasibility of using granular media filtration to reduce the radium activity levels in the radium removal treatment pond effluent.
	Gulf Minerals Ltd.	Have undertaken screening and pilot scale studies to reduce the Ra-226 activity from the treatment pond effluent to less than 10 pCi/l total using filtration processes. Full scale studies are proposed using granular media filtration.

Research Category	Research Organization(s)	Brief Description of Research
<p>Mill Wastewater - Radium Removal (from Tailings Area Effluents) (cont'd.)</p>	<p>University of Calgary</p>	<p>The suitability of manganese impregnated acrylic fibres for radium and thorium removal has been investigated in laboratory and field tests and the technique shows promise. Much of the work thus far has been directed towards analytical techniques.</p>
<p>BaRaSO₄ Sludges (Characterization and Stability)</p>	<p>CANMET</p>	<p>Commissioned, in 1978, a survey and critical assessment of the literature on the removal of radionuclides from process streams.</p>
<p>BaRaSO₄ Sludges (Characterization and Stability)</p>	<p>CANMET</p>	<p>Commissioned, in 1978, a project to document the chemical, physical and radiological characteristics of the existing sludges at Elliot Lake.</p>
<p>BaRaSO₄ Sludges (Characterization and Stability)</p>	<p>CANMET and the University of Calgary</p>	<p>Conducted studies on the chemical, physical and biological stability of sludges.</p>

Research Category	Research Organization(s)	Brief Description of Research
Tailings Management	Eldorado Nuclear Ltd.	<p>Eldorado's Beaverlodge operation use mill tailings as backfill in the mine. Studies were undertaken to examine the use of mill tailings as backfill in a mine. They have recently undertaken a project to refine the current cycloning technique used to separate the tailings used for mine backfill. The objective is to reduce the quantity of slimes discharged to the tailings area and thus use mine backfilling as a "wastemanagement technique".</p>
	Denison Mines Ltd.	<p>The feasibility of using a portion of the tailings underground is being examined. Studies have been underway for several years and are continuing.</p>
	Denison Mines Ltd. and CANMET	<p>Conducted laboratory and pilot scale studies to determine the feasibility of removing pyrite and possibly radionuclides from the tailings prior to discharge to the tailings impoundment.</p>
	Rio Algom Ltd. and CANMET	<p>In 1975, at the Quirke Mine, a series of test pits were filled with tailings and the effects of various surface treatments were examined. One of these included flooding one pit to a depth of five feet of water. This study is continuing.</p>

Research Category	Research Organization(s)	Brief Description of Research
Tailings Stabilization	Rio Algom Ltd.	Undertook research and field scale trials on vegetation of abandoned tailings areas beginning in 1970 and continuing to the present time.
	University of Guelph and Rio Algom Ltd.	Initiated in late 1969 laboratory scale studies to determine possible techniques and plants suitable for the revegetation of the Elliot Lake tailings. Examined, as an aid, utilization of composted soft wood, municipal garbage wastes and sewage sludge.
	CANMET (In collaboration with Rio Algom Ltd. and Denison Mines Ltd. for	Undertook a comprehensive program to produce a self-sustaining vegetative cover on inactive tailings areas. Basic laboratory studies were initiated in 1971. These were expanded to include small field plots and eventually to large scale studies covering many acres. Examined utilization of various grass species, trees, uptake of radionuclides, leaching rates, and undertook other work to identify and characterize the acid-producing mechanisms which operate within the tailings area. These programs are continuing.

Research Category	Research Organization(s)	Brief Description of Research
Tailings Stabilization (cont'd.)		Undertook at Laboratory scale, a lysimeter study (one year) to assess the release of acid and radionuclides from pyrite free tailings under chemical and biological oxidizing conditions.
		Undertook a feasibility study on the composting of sawdust with a view to adding this material to the tailings surface layer (i.e., approximately the top six inches) to facilitate the growth of vegetation.
	Eldorado Nuclear Ltd.	Undertook preliminary studies on vegetating tailings areas at Beaverlodge, Sask.
	Carleton University	Undertook laboratory scale leaching studies on tailings from the Beaverlodge Mine to define the operative reaction mechanism(s).

Research Category	Research Organization(s)	Brief Description of Research
Tailings Stabilization (cont'd.)	University of Toronto	Recently completed laboratory scale studies with the objective of quantifying and understanding the leaching process of Ra-226 from tailings at an active and an inactive site.
	Eldorado Nuclear Ltd.	Commissioned feasibility studies on pelletizing tailings from Beaverlodge.

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