

TECHNOLOGY PERSPECTIVES ON THE REMOVAL OF RADIOSTRONTIUM FROM LARGE VOLUMES OF GROUNDWATER

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ABSTRACT

Radiostrontium (^{90}Sr) is a relatively long-lived radionuclide commonly found in many groundwater systems migrating from historic nuclear waste storage sites. Numerous approaches have been developed for the remediation of such contaminated ground waters. Several strontium-removal technologies have been applied/assessed at Chalk River Laboratories (CRL) site, Atomic Energy of Canada Limited (AECL). These include simple ion exchange, (using non-selective natural zeolites or selective synthetic inorganic media) and precipitation/filtration with ion exchange polishing.

The challenges for the remediation of ^{90}Sr -contaminated waters arise from the need to achieve very high removal efficiencies from groundwater with relatively high concentrations of other inactive cations. Low-cost natural zeolites are non-selective for strontium over other divalent cations, and competition for sorption sites by such ions (notably calcium) results in very limited ^{90}Sr removal before effluent concentrations exceed discharge objectives. This produces large volumes of solid waste. Higher removal efficiencies, and much smaller volumes of solid waste, are obtained with strontium-specific synthetic ion exchangers, but the cost of these innovative sorbents is prohibitive for all but limited small-scale applications unless the cost of waste disposal are extremely high. In certain applications, non-selective treatment involving bulk chemical precipitation and filtration may be a more viable option. Some groundwater treatment operations at the CRL site have led to the application of AECL's CHEMICTM (CHEmical treatment and MICrofiltration) process in combination with final effluent polishing using ion exchange to achieve discharge targets while maintaining moderate cost and waste volumes. This paper describes key treatment approaches to meet technical, environmental and economic targets for remediation of large volumes of groundwater containing low concentrations of strontium.

INTRODUCTION

There are several critical performance aspects to be considered when selecting a suitable technology for treating radiostrontium-contaminated waters. The main consideration for retaining a technology is its ability to produce high quality discharge water while minimizing the volume of secondary waste that requires long-term management. However, life-cycle cost that includes capital, operating and waste management costs must also be taken into account. The magnitude of the volume and characteristics of the contaminated water to be treated will also influence the choice of technology applied. Typically, ^{90}Sr -contaminated ground waters consist of large volume ^{90}Sr -bearing plumes that would require many years of continuous treatment. These waters will contain commonly occurring species such as calcium, iron, magnesium, manganese, sodium, and potassium. Calcium is of special interest because of its close chemical

similarity to strontium. If present in high enough concentration it can interfere with certain strontium removal process (e.g., direct ion-exchange) and force the selection to other processes where strontium is either selectively remediated or is removed in parallel with calcium. It should be also noted that although the goal is to remove low concentrations of ^{90}Sr (a few hundreds to a few thousands Bq/L) with high removal efficiencies, the magnitude of total strontium (Sr) concentration in the groundwater is generally dictated by the presence of natural, non-radioactive strontium (^{87}Sr). Generally, the inactive strontium and ^{90}Sr concentrations in the ground water are in the order of parts-per-million and parts-per-trillion, respectively.

Strontium Removal Technologies

Removal technologies for ^{90}Sr (or total Sr) can be categorized as selective or non-selective. Generally, non-selective technologies indiscriminately remove the bulk of ionic species from waters under a wide range of conditions. These technologies include ion exchange using natural zeolite or organic ion-exchange resins, chemical precipitation, evaporation and reverse osmosis (RO) among others (Fig. 1).

The non-selective processes, although flexible/versatile and suitable for treatment of dilute waste solutions, become less effective in the presence of large concentrations of dissolved metals. For example, the high levels of total dissolved solids (TDS) present in many contaminated ground waters results in high solution conductivity, which can cause rapid exhaustion of ion-exchange media requiring early regeneration or removal, thus producing higher amounts of secondary wastes. High conductivity waters may also cause other problems such as RO membrane fouling and evaporator scaling when the concentrations of scale-forming species exceed their solubility limits during contaminant removal or enrichment. To avoid these problems, pre-treatment of the waters is essential; otherwise the RO or evaporator units must be operated to lower waste-volume reductions (ratio of waste volume treated to volume of concentrate) thus producing higher secondary-waste volumes.

Previous work at AECL has demonstrated that contaminants can be effectively sequestered from higher conductivity ground waters by applying chemical precipitation techniques [1,2,3,4,5]. For example, the presence of high concentrations of species such as iron, calcium, and magnesium would enhance strontium removal through co-precipitation, and sorption on the metal hydroxide and metal carbonate precipitates. In recent years, advances and the commercial introduction of certain selective-sorption media for strontium removal from waste waters has re-opened the potential to apply such technologies as alternatives to conventional non-selective technologies based on ion-exchange media involving organic resins or natural zeolites (e.g., clinoptilolite, chabazite). Sorption media such as titanates (e.g., SrTreat), synthetic zeolites or crystalline silicotitanate, CST (e.g., IONSIV-911) have been employed to discriminately separate strontium from other cations resulting in higher removal efficiency and lower volumes of secondary waste [6,7,8,9,10]. However, these sorbents, engineered specifically for applications involving high conductivity contaminated-waters, have seen only limited applications.

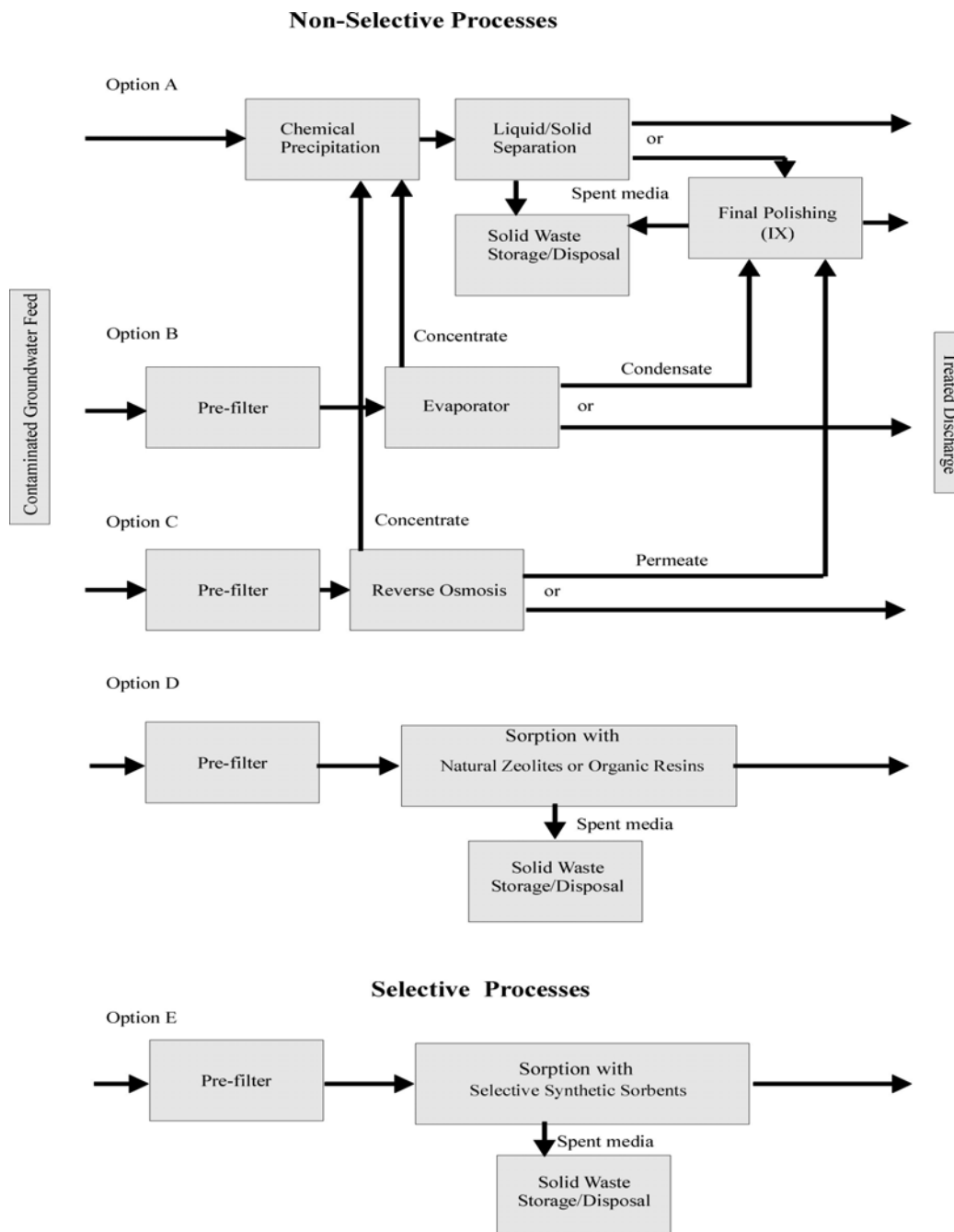


Fig. 1. Technology options for ⁹⁰Sr removal from contaminated groundwater.

TREATMENT APPROACH

Among the different approaches for removing ⁹⁰Sr from waters, we have selected three treatment approaches for evaluation in the current study. In the first approach, strontium removal using fixed-bed ion-exchange with natural zeolite media was retained as the reference technology for comparison because of its simplicity, versatility and low cost of the media. Secondary waste produced as spent zeolite can be easily stabilized in suitable waste forms that are well

characterized for long term storage and final disposal. There is relevant experience in the implementation and operating of these systems at AECL and elsewhere [3,4,5,6,7]. In the second approach, we have evaluated two commercially-available synthetic sorbents for their known selectivity for strontium-removal [10,11] over non-selective natural zeolites. Finally, we have selected and assessed against the first two approaches AECL's CHEMIC process based on non-selective precipitation, followed by solid-liquid separation with or without ion-exchange polishing.

Groundwater Feed Characteristics

For the present study, we tested two ground waters at the CRL site to illustrate the feasibility of applying selective versus non-selective strontium-removal approaches. The first stream (SW) contained relatively high levels of calcium (25 mg/L) and the other groundwater stream (CP) contained low levels of calcium (2 mg/L). The approximate composition of each groundwater is listed in Table I.

Table I Groundwater Characteristics^a

Species	Concentration	
	SW Groundwater (avg.)	CP Groundwater (avg.)
Strontium (natural Sr) (mg/L)	0.2	0.01
Strontium-90 (Bq/L)	1700	1500
Strontium-90 (mg/L)	3.34×10^{-7}	2.95×10^{-7}
Calcium (mg/L)	25	2
Iron (mg/L)	0.01	0.05
Magnesium (mg/L)	10	0.7
Sodium (mg/L)	100	12
Potassium (mg/L)	4	0.9
Condcutivity (μ S/cm)	800	60
Solution pH	6.5	6.0

^a Although the concentrations for these species will have large variability with time, the values reported are average of several groundwater samples that have been bulked for testing.

Process Testing

The equipment layout was identical for sorption tests using either natural zeolites or selective-sorbent media. The ⁹⁰Sr-contaminated groundwater was pumped through the media packed in a fixed column configuration. The effluent from the sorbent bed was collected and monitored for contaminant levels. The majority of the test runs continued well above the arbitrary discharge target level of 50 Bq ⁹⁰Sr/L.

To test the CHEMIC method, strontium was removed from solution along with calcium through an alkaline carbonate precipitation step, followed by microfiltration (referred to as the partial CHEMIC). The filtered solution was introduced to a polishing step based on sorption to achieve

lower discharge targets, as necessary. The combined precipitation and sorption steps are referred to as the full CHEMIC process.

Sorption Tests

Laboratory-scale experiments were performed using one centimetre diameter glass columns packed with either natural zeolites (clinoptilolite, SteelHead Specialty Minerals TSM 140; chabazite, TSM 310) or selective synthetic media (UOP, IONSIV IE-911 (CST); Selion OY, SrTreat, a modified titanium oxide). The media were slurried into the columns to an average bed height of 10 centimeters (8 milliliter total bed volume, BV). Several volumes of distilled water were flushed through the columns prior to the beginning of each test in an attempt to remove fines and avoid column plugging. The CST media was not pre-washed in the present tests. The laboratory sorption columns were fitted with glass frit as filters. Using a peristaltic pump, water collected from the contaminated stream was passed through the packed column at a constant flow of 4 mL/min or 0.5 BV/min. This flow rate was within the range commonly used to meet residence time and column pressure-drop considerations. Effluent from the column was collected at regular intervals using a fraction collector, and ^{90}Sr concentration determined by liquid scintillation counting. The removal of other non-radioactive species (Ca, Mg, K, Na) was monitored by analyzing samples by inductively coupled plasma atomic emission spectroscopy and/or atomic absorption spectroscopy. Data from the operation of a groundwater treatment facility at CRL utilizing natural-zeolite columns (25 liters BV, fitted with fibreglass packing as filter) at flow rates (0.2 –0.4 BV/min), similar to the laboratory tests, were also included in the study.

CHEMIC Tests

The CHEMIC process was tested in a batch mode. The groundwater feed samples, in stirred vessels, were pH adjusted to 10.5 using sodium hydroxide followed by the addition of carbonate (as sodium carbonate) in a 2:1 molar ratio to the calcium concentration. After approximately 1 hour of precipitation time, the dilute slurry was filtered using a 0.2-micrometer average pore-diameter filter. The filtrate was collected and analyzed for ^{90}Sr . In some tests, the collected filtrate was passed through a pre-selected sorption/ion exchange column as a final polishing step to further reduce the ^{90}Sr concentration.

RESULTS AND DISCUSSIONS

Strontium Removal by Natural Zeolite Sorption (Approach 1)

Removal performance data for ^{90}Sr using two natural zeolites were determined by following the ^{90}Sr breakthrough trends. Tests with the SW groundwater revealed that between the two natural zeolites tested, chabazite performed better. As illustrated in Fig. 2, a ^{90}Sr breakthrough concentration of less than 50 Bq/L (arbitrary target) was maintained for nearly 450 BV with chabazite whereas clinoptilolite gave 120 BVs. The early breakthrough of calcium in the SW water explains the rapid exhaustion of the sorbent bed and the subsequent early Sr elution from the column. This contrasts with the experience gained from processing over 20 millions liters of low calcium CP water. Operating either clinoptilolite or chabazite columns with the CP stream at

an average flow rate of 0.3 BV/min, several thousand BVs (2000 to 4000 BV) could be processed before exceeding ^{90}Sr discharge values in the range of 5 to 60 Bq/L. Results from a recent performance evaluation of a clinoptilolite column operating with CP water showed ^{90}Sr discharge levels of 25 Bq/L after treating over 4000 bed volumes (Fig. 2).

The difference in ^{90}Sr removal efficiencies for the two streams can be attributed to the higher conductivity and calcium content of the SW water and natural zeolites low selectivity for strontium over these species. While the natural zeolite sorbents were more than adequate for ^{90}Sr removal of CP type contaminated waters, if operated with SW water, large volumes of spent zeolite secondary waste would obviously be generated to meet the same discharge target. The need for a more selective sorbent for the SW type water is apparent if a sorption-based technology is to be selected.

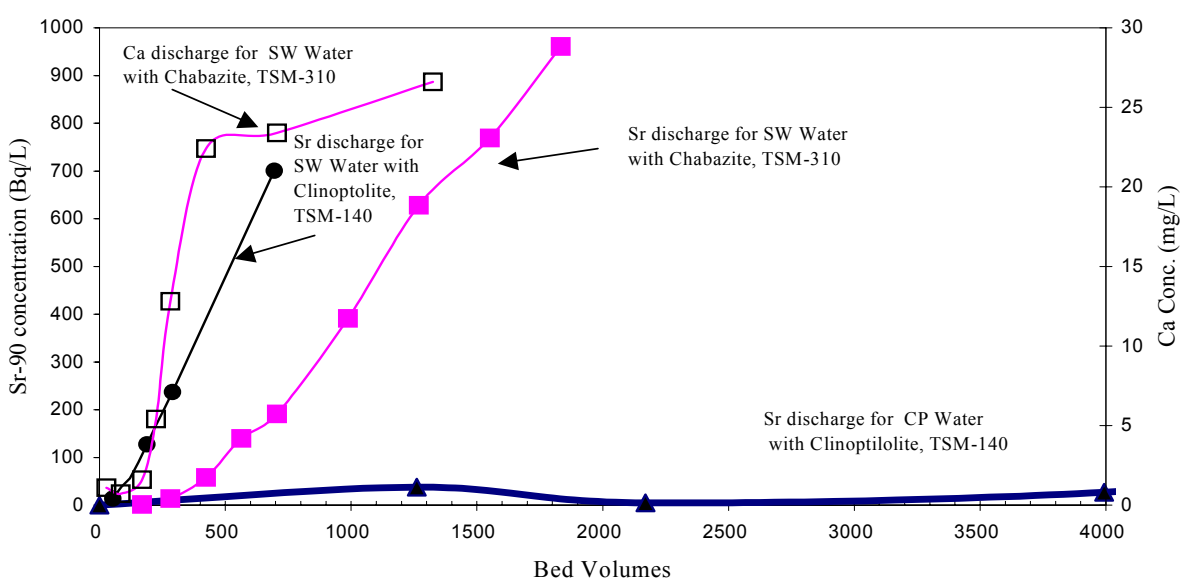


Fig. 2. Breakthrough curves for strontium-90 using natural zeolites for SW and CP ground waters.

Strontium Removal by Sr-Selective Sorbent (Approach 2)

Results from laboratory-scale sorption tests for ^{90}Sr and calcium breakthrough behavior with two selective sorbents, CST and SrTreat for SW waters, are given in Fig. 3.

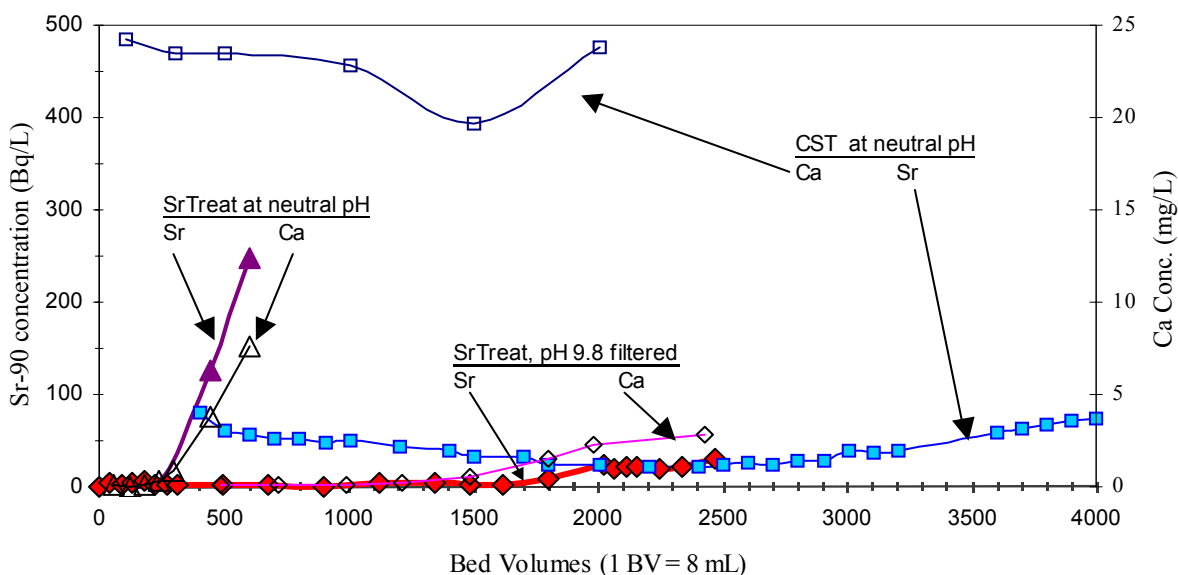


Fig. 3. Breakthrough curves for strontium-90 and calcium using SW groundwater with various synthetic sorbents.

The CST material, known for its selectivity for ^{90}Sr over competing calcium [7,8,9], showed a strontium-removal performance of approximately 7.5-times higher than that through sorption by chabazite. After initially high discharge concentrations of ^{90}Sr (around 80 Bq/L) from the column, the target level of 50 Bq/L could be maintained for up to 3500 bed volumes of CST. As expected, within the experimental and analysis errors for calcium, CST did not retain calcium and the discharge concentration remained essentially at the feed concentration levels while ^{90}Sr was being selectively removed. The initial 80 Bq/L, higher than the discharge target of 50 Bq/L for ^{90}Sr , is attributed to fine-particle leakage, commonly observed with CST columns. To eliminate this effect extensive pre-washing of the column with deionized water at the operating pH of the column will be necessary.

Results with SrTreat, under similar conditions, showed an initial ^{90}Sr breakthrough around 30 Bq/L after 300 BV had passed through the column. The ^{90}Sr discharge concentration from the column rapidly reached a level above 120 Bq/L after 450 BVs. SrTreat exhibited poor selectivity for strontium over calcium as indicated by calcium's early removal followed by a rapid increase in its discharge concentration, matching the ^{90}Sr breakthrough. As SrTreat is recommended for more alkaline applications, the pH of the contaminated water, originally at neutral, was adjusted to 9.8 and further sorption tests were conducted. Adjusting the pH of the solution had the effect of precipitating some of the metals (e.g., Mg) as hydroxide and some calcium as carbonate by absorption of atmospheric CO_2 . After filtering the solid precipitates, calcium concentration in solution was found to be reduced from 25 to 11 ppm and ^{90}Sr level fell from 1700 Bq/L to 800 Bq/L. The decrease in ^{90}Sr value can be attributed to removal by sorption on to the solid precipitates. When the SrTreat sorption test was carried out using the filtered water after pH

adjustment, the column operated past 2500 BVs at which point ^{90}Sr level had reached 25 Bq/L. The relatively poor ^{90}Sr removal efficiency at neutral pH appears to be because SrTreat is not totally selective for strontium over calcium, despite its reported high capacity and selectivity for strontium in high conductivity waters [8].

Strontium Removal by CHEMIC Process (Approach 3)

Laboratory-scale test results using the precipitation-microfiltration process performed on SW water are described in this section. The partial CHEMIC process uses chemical precipitation followed by micro-filtration to remove ^{90}Sr from contaminated waters. The full CHEMIC process uses the partial CHEMIC followed by a polishing sorption step to produce high quality discharge waters. The partial and full CHEMIC processes were applied to the SW groundwater to assess its effectiveness for ^{90}Sr removal as compared to treatment based on direct sorption system. The test results are given in Table II and Fig. 4.

Table II Results from Applying AECL's CHEMIC Process on SW Water
(Average [Ca] = 25 mg/L; Average [^{90}Sr] = 1700 Bq/L)

SW Feed Volume	Avg. Temp. (°C)	CHEMIC Precipitation	CHEMIC Filtrate (after precipitation-microfiltration)			Final Treated water (after Precipitation-microfiltration and sorption) ^a
			Ca (mg/L)	^{90}Sr (Bq/L)	Cond. (mS/cm)	
		pH				^{90}Sr (Bq/L)
18.0 L	21	>12	2.20	751	-	63 Bq/L @ 100 Bed Volumes (BV) with chabazite
18.0 L	21	>12	2.1	740	4.33	5 Bq/L @ 3900 BV with CST
18.0 L	21	10.7	4.7	24	1.23	4 Bq/L @ 1300 BV with chabazite

^a After precipitation and filtration the filtrate was neutralized with CO_2 prior to sorption tests.

As the reaction occurs under alkaline conditions, the precipitation tests were performed with the SW groundwater at pH values greater than 12. Results from these tests produced ^{90}Sr concentrations around 750 Bq/L and calcium in the range of 2-3 mg/L in the filtrate after microfiltration of the precipitates. A chabazite sorption test was conducted with the filtrate with low calcium content from the CHEMIC test to determine if it was possible to use a chabazite column as the polishing step in the CHEMIC process train. The filtrate from the precipitation step was therefore passed through a chabazite column and the results showed that the chabazite column was exhausted much more quickly than anticipated (>50Bq/L @ < 100 Bed volumes) even with lower calcium concentrations. The poor results were attributed to zeolite's relatively lower cation-exchange capacity for high conductivity solutions, caused in particular when caustic was used for pH adjustment in the precipitation step.

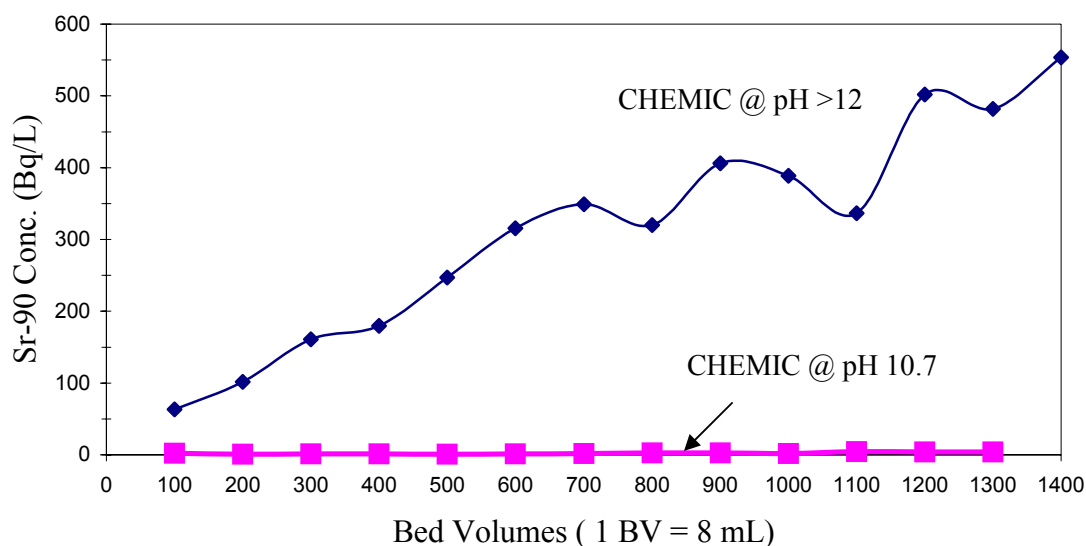


Fig. 4 Performance of filtrate (obtained from CHEMIC precipitation-microfiltration) treatment by chabazite sorption at different precipitation pH values using the SW water.

To improve the performance of zeolite after the precipitation step, subsequent CHEMIC precipitation tests were conducted at a lower pH of 10 to 10.7, which gave reduced sodium concentrations (and hence lower conductivity values) to more acceptable levels. Results from the precipitation step showed a marked improvement in ^{90}Sr removal. The filtrate now contained only 25 Bq ^{90}Sr /L. As strontium removal in the CHEMIC process is partially achieved through sorption on the precipitated solids, it is possible that higher sodium concentrations at high pH may have been hindering the relative removal of ^{90}Sr ions by sorption. The ^{90}Sr results from the subsequent chabazite-polishing step also showed expected improvements (less than 5 Bq/L ^{90}Sr in the final effluent for greater than 1300 bed volumes) from the reduced sodium concentration at the lower pH values employed in the tests.

Previous experience (unpublished data) at AECL with the application of the CHEMIC process to the CP water produced discharge water with ^{90}Sr concentrations in the range of 30 to 200 Bq/L. However, the treatment required the addition of calcium to the feed water to raise the concentration from 2 ppm to above 50 ppm to permit the chemical precipitation to proceed (above the solubility of CaCO_3) and to provide sufficient mass for effective filtration. Since secondary waste (separated precipitate) volume is relatively high with this approach and direct sorption with non-selective sorbents such as natural zeolites is effective, the CHEMIC process would not be the preferred approach for strontium removal from waters containing very low calcium concentrations.

COSTS FOR REAGENTS AND SECONDARY WASTE MANAGEMENT

In an attempt to develop a guideline for selecting the treatment approach from among the two sorption and CHEMIC approaches for the SW (high calcium) and CP (low calcium) type ground waters, we have used the ^{90}Sr -removal performance data and estimated annual costs of reagents and secondary-waste management. The results are summarized in Table III.

The key assumptions involved in this analysis include:

- the operating costs of different approaches excluding the reagent and secondary-waste management costs are similar for the different treatment approaches;
- the annualized capital cost (including site preparation, licensing, water collection and building) for the treatment plant over the life of the plant (e.g., 15 years) is comparable among the different treatment approaches. It should be noted that the portion of the capital cost attributable to the processing equipment in the CHEMIC plant would be higher than that in the sorption-based treatment plant. However, the corresponding annualized capital cost for the CHEMIC equipment would be only incrementally high and would not make a significant increase in the overall annualized capital cost distributed over the plant life.
- the operating costs related to ground-water collection, chemical and radiochemical analysis, overall labor and utility are similar among the different approaches; and
- the CHEMIC approach is not technically appropriate for ground waters with low calcium concentrations (approximately 2 mg/L) and low conductivities.

The simplified cost analysis suggests that even for high calcium type ground waters, the use of selective sorbents such as the CST is not the preferred approach. High reagent costs make this choice unappealing despite the generation of low volumes of secondary wastes. The direct sorption by natural zeolite provides the next lowest combined costs for reagent and secondary-waste management. Depending on the treated water target, the application of partial or full CHEMIC with natural-zeolite-sorption step produces relatively low secondary-waste volumes, and lowest combined costs for reagents and secondary-waste management.

The CHEMIC process is not appropriate for the low calcium content ground waters. The direct-sorption approach with natural zeolite appears to be the preferred approach since it provides low combined costs for reagents and secondary-waste disposal.

Table III Summary of Annual Reagent Cost versus Strontium Removal Performance by Different Technologies ^{a,b,c}

Option Description	Groundwater Stream Feed Conc.	⁹⁰ Sr in Treated Water (Bq/L)	Waste Volume Reduction Factor (approx.)	Quantity of Reagents Needed	Projected Reagent Cost ^{b,c} (approx. \$/a)	Projected Secondary -Waste Disposal Cost ^d (\$/a)	Projected Total Cost (Reagent and Waste Disposal) (\$/a)
Non-selective Sorption with Chabazite	SW ⁹⁰ Sr = 1700 Bq/L Ca = 25 mg/L Cond. = 800 μ S/cm	<50	650 (or 29 m ³ secondary waste)	29 m ³	160 000	290 000	450 000
Selective Sorption with CST	SW	<50	4800 (or 3.9 m ³ secondary waste)	3.9 m ³	740 000	39 000	779 000
Partial CHEMIC (without sorption step)	SW	<50	5400 (or 3.5 m ³ secondary waste)	800 kg NaOH + 16 000 kg Na ₂ CO ₃	22 400	35 000	57 400
Full CHEMIC with Chabazite Sorption	SW	<5	3400 (or 5.6 m ³ secondary waste)	800 kg NaOH + 16 000 kg Na ₂ CO ₃ + 2.1 m ³ chabazite	34 000	56 000	90 000
Non-selective Sorption with Chabazite	CP ⁹⁰ Sr = 1500 Bq/L Ca = 2 mg/L Cond. = 60 μ S/cm	<50	4800 (or 4 m ³ secondary waste)	4.0 m ³ chabazite	22 000	40 000	62 000

^a Reference capacity: 38 L/min (10 US gpm) Plant; the chemical reagent cost and secondary waste-volume would increase proportionately with the rate at which water is treated; total volume of contaminated water treated per annum = 19 million litres.

^b The cost estimation was based on US\$ 189.20/kg for CST and US\$ 5.50/kg for chabazite. The density of the sorbent is 1.0 kg/L.

^c The cost for NaOH is \$1.7/kg and \$1.3/kg for Na₂CO₃.

^d Assumed disposal cost for low-level radioactive secondary waste, US\$10 000 /m³ of waste.

CONCLUSIONS

On the basis of radiostrontium removal results obtained for high calcium and low calcium content ground waters using direct sorption treatment (involving selective or non-selective

sorbents) and the CHEMIC process (involving precipitation-microfiltration with or without a sorption-based polishing step), the following conclusions can be drawn:

- Direct sorption tests using non-selective natural zeolites for strontium removal from waters with high levels of calcium and high conductivity, have shown that large volumes of spent zeolite (secondary waste) would be generated to achieve low Sr discharge limits, rendering this treatment approach unattractive.
- Direct sorption of ^{90}Sr from high calcium content groundwater by selective sorbents such as crystalline silicotitanate is technically feasible to provide low ^{90}Sr concentrations in the treated water. The use of crystalline silicotitanate would provide greater strontium-removal capacity than chabazite by a factor of 7.5 while consuming significantly less sorbent. However, the high cost of crystalline silicotitanate negates the cost saving benefits derived from the disposal of low amounts of secondary waste.
- SrTreat sorbent would not be suitable for use in direct sorption-based treatment of neutral pH waters with high calcium concentrations.
- The experience with the application of the Full CHEMIC process, involving chemical precipitation and microfiltration and a final polishing step with chabazite sorption, to the high calcium content groundwater shows that this approach can readily produce low ^{90}Sr concentrations in the treated water.
- Since the majority of calcium has been removed by precipitation in the Full CHEMIC process, there is no apparent advantage for using high cost sorbents, such as crystalline silicotitanate or SrTreat, over natural zeolite sorbents.
- For low calcium content ground waters with relatively low conductivity, CHEMIC process is not the preferred option.
- The application of non-selective sorbents such as natural zeolites for the removal of radiostrontium from low calcium content ground waters is the preferred approach.

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