On-Site Pilot Study - Removal of Uranium, Radium-226 and Arsenic from Impacted Leachate by Reverse Osmosis – 13155

McMurray, Allan[a]. Everest, Chris[a]. Vandergaast, Gary[b]. LaMonica, David[c]. Rilling, Ken[a].

[a]Conestoga-Rovers and Associates, 651 Colby Dr, Waterloo, ON, Canada

[b]Atomic Energy of Canada Ltd, 115 Toronto Road, Port Hope, ON, Canada

[c]RoChem Membrane Systems Inc., 430 30th Street, Hermosa Beach, CA, USA

ABSTRACT

Conestoga-Rovers & Associates (CRA-LTD) performed an on-site pilot study at the Welcome Waste Management Facility in Port Hope, Ontario, Canada, to evaluate the effectiveness of a unique leachate treatment process for the removal of radioactive contaminants from leachate impacted by low-level radioactive waste. Results from the study also provided the parameters needed for the design of the CRA-LTD full scale leachate treatment process design. The final effluent water quality discharged from the process to meet the local surface water discharge criteria. A statistical software package was utilized to obtain the analysis of variance (ANOVA) for the results from design of experiment applied to determine the effect of the evaluated factors on the measured responses. The factors considered in the study were: percent of reverse osmosis permeate water recovery, influent coagulant dosage, and influent total dissolved solids (TDS) dosage. The measured responses evaluated were: operating time, average specific flux, and rejection of radioactive contaminants along with other elements. The ANOVA for the design of experiment results revealed that the operating time is affected by the percent water recovery to be achieved and the flocculent dosage over the range studied. The average specific flux and rejection for the radioactive contaminants were not affected by the factors evaluated over the range studied. The 3 month long on-site pilot testing on the impacted leachate revealed that the CRA-LTD leachate treatment process was robust and produced an effluent water quality that met the surface water discharge criteria mandated by the Canadian Nuclear Safety Commission and the local municipality.

INTRODUCTION

Conestoga-Rovers and Associates Limited (CRA-LTD) was retained by the Atomic Energy of Canada Limited (AECL) to provide a design for a full-scale treatment plant for the removal of contaminants of concern (COC) from impacted leachate at the Welcome Waste Management Facility (WMF), an existing low-level radioactive waste site located in Port Hope, Ontario. The main COC are uranium (U), radium-226 (Ra-226), and arsenic (As). The effluent quality objectives for the full-scale plant include: a high level of assurance for human health, a reduction of radiological discharge in discharge water to levels as low as reasonably achievable, and minimization of the total treated leachate loading to Lake Ontario. CRA-LTD concluded from its bench scale test results that the chemical pre-treatment of the leachate followed by clarification then concentration of the COC with the Rochem Spacer Tube (ST) reverse osmosis (RO) membrane module as potentially feasible [1]. Radioactive contaminants have been effectively concentrated by RO in leachate pre-treatment [2, 3, 4, 5, 6], drinking water pre-treatment [7], and radioactive waste processing [8]. The RO technology was identified as the best demonstrated technology for removal of radium-226, radium-228, and uranium from leachate by the United States of America Environmental Protection Agency (USEPA) [9]. The focus herein is of the on-site pilot study performed by CRA-LTD, which utilized a fractional factorial statistical design of experiment (SDOEX) to evaluate the identified approach on a pilot scale as well as determine if the approach is practical and robust in removing and concentrating COC. Additional objectives included determining the operating envelope of the Rochem ST RO modules, the impact of recovery of Rochem ST RO module on performance, chemical pre-treatment and influent quality on module performance, and design parameters needed for the full-scale plant design. These objectives were identified by the USEPA as being important in assessing the overall effectiveness of a treatment technology for the management of radioactive residuals from drinking water [10].

DESCRIPTION

The on-site pilot testing was performed for three consecutive months. The pilot system was contained in a 2.4 m by 12.2 m metal wind/water tight tunnel style containerized intermodal freight container; referred to as the "pilot trailer". The aerial photo in Figure 1 is of the Welcome WMF site in Port Hope with the location of the pilot trailer relative to the existing leachate treatment building shown. The pilot system was comprised of a mixing and storage tank, chemical metering equipment, an inclined plate clarifier, a clarifier supernatant storage tank, and the Rochem RO-510 PT2 pilot unit with permeate and concentrate storage tanks. The process flow diagram of the pilot system is shown in Figure 2. The photos displayed in Figure 3a and 3b are of the chemical pre-treatment equipment and pilot Rochem RO-510 PT2 unit, respectively, in the pilot trailer.

The study applied a fractional factorial SDOEX to determine the influence of the factors: percent recovery, pre-treatment, and TDS spike. The factor percent recovery refers to the percentage of the total permeate flow to that of the influent flow to the first RO module. The levels evaluated were: 75%, 80%, and 85%. The factor pre-treatment refers to the ferric chloride dosage levels applied in the clarifier; ferric chloride applied as a flocculating agent. The levels evaluated were: pre-treatment (ferric chloride dosage of 130 ppm) and no pre-treatment (no ferric chloride added). The levels evaluated for the TDS spike factor were: TDS spike (increased electrical conductivity of first RO module influent between 1,000 and 1,200 uS/cm) and no TDS spike (unaltered influent electrical conductivity to the first RO module). The design matrix of the SDOEX involved nine runs the factor levels for each of the runs are provided in Table I. This design was applied to minimize the number of runs, evaluate main effects, and evaluate error. The conditions of run 2 were replicated in run 3 and 8 to obtain a measure of error. The chemical analysis was performed by a laboratory accredited by the Canadian Association for Environmental Analytical Laboratories.

Fig. 1: Aerial photo of the Welcome Waste Management Facility in Port Hope Ontario Canada. Identified in the photo are the locations of the pilot trailer (2.4 m by 12 m metal wind/water tight tunnel style containerized intermodal freight container containing pilot equipment utilized in study), and existing leachate treatment building. The leachate is collected in two main collection ponds; south east collection pond shown in the photo. Also, identified in the photo are the locations of the leachate water supply line, wastewater return line from pilot trailer, and power supply line to pilot trailer.

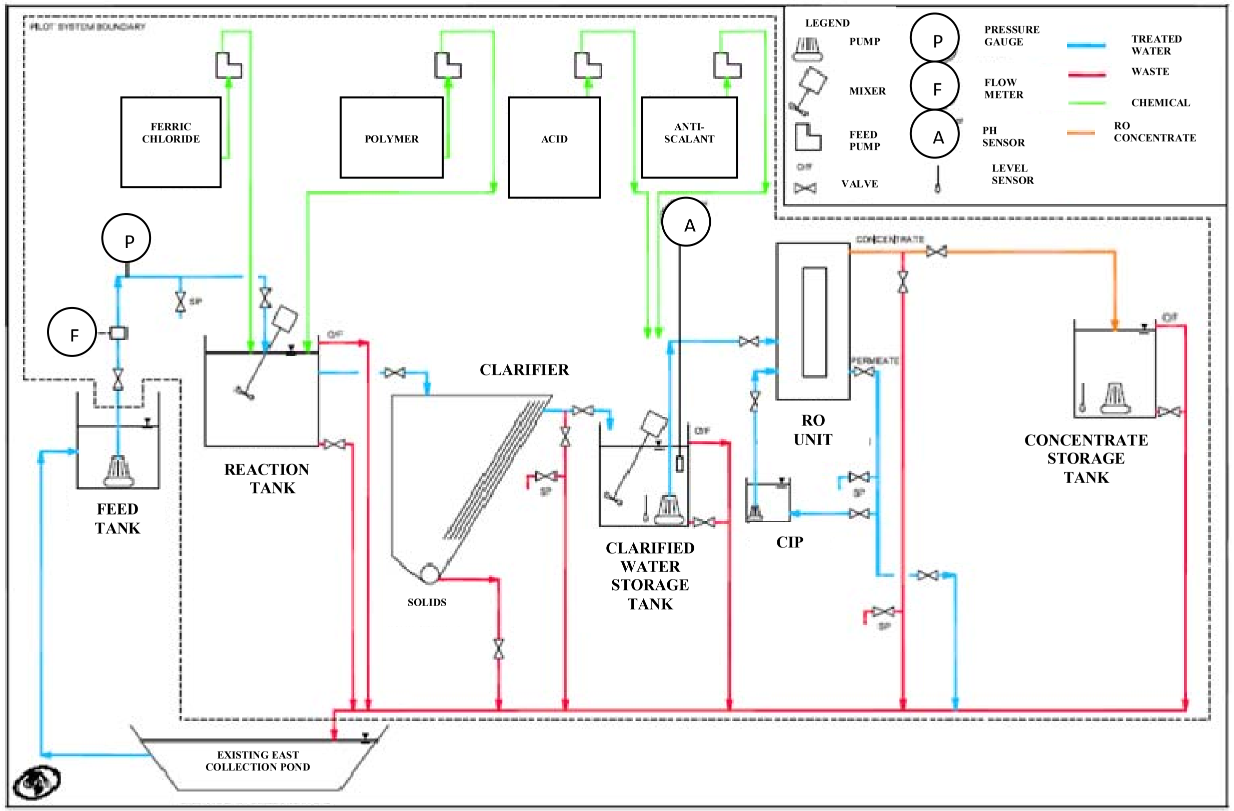

Fig. 2: Process flow diagram of the on-site pilot test system utilized at the Welcome Waste Management Facility in Port Hope Ontario Canada. The pilot system was comprised of a mixing and storage tank, chemical metering equipment, an inclined plate clarifier, a clarifier supernatant storage tank, concentrate storage tank and the Rochem RO-510 PT2 pilot unit. The influent leachate to the pilot system is drawn from the existing east collection pond. The liquid and solid (clarifier sludge) waste streams were directed back to the east collection pond.

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Fig. 3: Photos of inside the pilot trailer applied in on-site pilot study. Pilot trailer utilized a 2.4 m by 12.2 m metal wind/water tight tunnel style containerized intermodal freight container to house the pretreatment processes for pilot testing. Shown in view “a)” is the chemical treatment equipment. The view in “b)” is of the Rochem RO-510 PT2 unit with three (3) Rochem ST modules oriented vertically with the permeate storage tank in the background.

Table I
Design Matrix for Design of Experimental Model[i]
for the Port Hope On-Site Pilot Study Design

Run #	Main Effect Level		
	Percent Recovery[ii]	Pre-treatment[iii]	TDS Spike[iv]
1	85	No	No
2	80	No	No
3	80	No	No
4	80	No	Yes
5	80	Yes	No
6	85	Yes	No
7	85	No	Yes
8	80	No	No
9	75	No	No

[i] Design of Experiment (SDOEX): Conestoga-Rovers & Associates Limited (CRA-LTD) evaluated three main effects; the first order interactions and higher effects were considered part of the error term. The model utilized was a crossed three-way mixed unbalanced model design to evaluate data from nine runs with three repeats.

[ii] Percent Recovery refers to the percent the reverse osmosis (RO) permeate flow is of the RO influent flow to the first RO module. Levels of percent recovery evaluated were 75%, 80%, and 85%.

[iii] Pre-treatment refers to the ferric chloride dosage level applied in the clarifier unit upstream of the RO unit to promote flocculation of suspended solids. Levels evaluated were pre-treatment (ferric chloride dosage at 130 ppm) and no pre-treatment (no ferric chloride added).

[iv] TDS Spike refers to injecting collected concentrate from the RO unit previous runs into the influent of the first RO module. The concentrate was added to increase the electrical conductivity in the range of 1,000 to 1,200 uS/cm. No TDS Spike means the electrical conductivity was not altered.

The RO concentrate was collected so that it could be utilized to increase the influent electrical conductivity of subsequent tests as required by the SDOEX. This electrical conductivity of 1,000 and 1,200 uS/cm is representative of the worst case scenario that the membranes would be exposed to when the full-scale plant is in operation to treat the leachate [11].

DISCUSSION

The SDOEX design matrix, shown in Table I, was performed to evaluate the effect of the stated factors on the measured responses: operating time, average specific flux, and rejection of COC, along with other elements. The operating time measured response refers to the average number of hours of operation of the RO system between cleaning cycles. The cleaning cycle is activated by the RO pilot system when the applied membrane pressure increases above 30% of the starting applied membrane pressure**.** The Stat-Ease Incorporated’s Design-Expert Version 8 software was utilized to generate the analysis of variance (ANOVA) for the crossed three-way mixed unbalanced model data. The first order and higher interactions were considered part of the error term in order to obtain the main effect of the factors on the measured responses in the test runs that could be accommodated in the allowed time frame. The results from the null-hypothesis tests of the ANOVA are provided in Table II.

Table II
Null Hypothesis Test Results of Main Effects in Regard to Factors Evaluated for Design of Experiment Model[i] for Port Hope On-Site Pilot Study

Measured Response	Main Effects	Significant[x]
Operating Time[ii]	Percent Recovery[vii]	Yes
	Pre-treatment[viii]	Yes
	TDS Spike[ix]	No
Average Specific Flux[iii]	Percent Recovery	No
	Pre-treatment	No
	TDS Spike	No
Uranium Rejection[iv]	Percent Recovery	No
	Pre-treatment	No
	TDS Spike	No
Radium-226 Rejection[v]	Percent Recovery	No
	Pre-treatment	No
	TDS Spike	No
Arsenic Rejection[vi]	Percent Recovery	No
	Pre-treatment	No
	TDS Spike	No

i Design of Experiment (SDOEX): CRA-LTD evaluated three main effects with first order interactions and higher considered as error. The SDOEX was a crossed three-way mixed unbalanced model design to evaluate data from nine runs with three repeats.

ii Operating Time refers to the hours of operation until cleaning is required. The operating conditions that activate the cleaning cycle are based on feed pressure in the first reverse osmosis (RO) module and decay in flux across the RO module.

iii Average Specific Flux refers to the flow per unit area across the RO module. Specific flux value is the flux corrected to a reference temperature and pressure.

iv Uranium Rejection refers to the percent of uranium (mass) retained in the concentrate from the RO module to that in the feed of the RO module.

v Radium-226 Rejection refers to the percent of radium-226 (radioactivity in becquerel) retained in the concentrate from the RO module to that in the feed of the RO module.

vi Arsenic Rejection refers to the percent of arsenic retained (mass) in the concentrate from the RO module to that in the feed of the RO module.

vii Percent Recovery refers to percent of permeate flow to that of the influent flow to the RO system on a volume basis.

viii Pre-treatment refers to ferric chloride dosage in the clarifier upstream of the RO unit. Levels evaluated were pre-treatment (ferric chloride dosage at 130 ppm) and no pre-treatment (no ferric chloride added).

ix TDS Spike refers to injecting collected concentrate from the RO unit previous runs into the influent of the first RO module. Concentrate was added to increase the electrical conductivity in the range of 1,000 to 1,200 uS/cm. No TDS Spike means the electrical conductivity was not altered.

x Significant refers to the result from the F-test at a 0.05% probability level. The main effect F-statistic was obtained from the ANOVA performed on the data for the design of experiment model.

In Table II the column titled “Significant” contains the results of the F-test against the F-distribution value at a 5% confidence level where “Yes” means that the F-statistic was considered significant based on the null-hypothesis test and is therefore assumed to influence the measured response. From the null hypothesis test results, shown in Table II, it was accepted that the operating time was affected by the percent recovery and pre-treatment, whereas the average specific flux and rejection of COC were not significantly affected by the factors evaluated over the range tested. The bar graph shown in Figure 4 is of operating time versus percent recovery for the data with no TDS spike. From this figure, it is noted that the highest operating time of 166 hours occurs at 80% recovery with pre-treatment. The operating times that occurred for 75% and 85% recovery were not significantly different; at approximately 80 hours. Thus at 80% recovery and pre-treatment the operating hours increases by at least 50% with respect to the other conditions evaluated. This is an important observation as maximizing operating time will minimize down time and overall chemical consumption leading to overall reduced operating costs. The profile of the bar graph profile for average specific flux versus percent recovery for data with no TDS spike shown in Figure 5 is relatively flat. This observation supports the null-hypothesis that the average specific flux response is independent of the factors evaluated over the range tested.

The percent rejection values shown in Table III are of the main COC for data with no TDS spike. The percent rejection values for uranium and arsenic were not significantly different for the pre-treatment and no pre-treatment test conditions. These results support the acceptance of the null-hypothesis test that pre-treatment, percent recovery and TDS spike do not significantly affect the percent rejection. The radium-226 percent rejection values for no pre-treatment were 50.0%, 90.0 and 91.7%. The percent rejection value of 50% can be considered an outlier. The radium-226 percent rejection values for the pre-treatment with no TDS spike test conditions were 75.0% and 83.3%. It may be inferred from these results that there is a difference in 226-radium rejection at the 2 conditions. The sensitivity to error in the analytical procedure for measuring the 226-radium radioactivity affects the standard deviation in the data and subsequently the null-hypothesis test. Arsenic has a relatively smaller atomic radius then that of radium-226 and the method of analysis for arsenic is less prone to error than that of radium-226. Based on these reasons and the results for uranium the null-hypothesis test result of radium-226 rejection not significantly affected by the factors over the range tested was accepted.

A deduction drawn from the results in Table II is the full-scale treatment system may provide tolerance to drift in the operating conditions without significantly impacting final effluent quality. This relative amount of tolerance in the system drift from the set point conditions should still result in an effluent water quality meeting the surface water quality discharge criteria mandated by the Canadian Nuclear Safety Commission. The application of ferric chloride at 130 ppm increases the RO system operating time which reduces membrane cleaning frequency and subsequently operator maintenance time. This should result in reduced overall operating costs for the process. The results of the on-site pilot testing demonstrated the efficacy of Rochem ST RO technology in removing COC from contaminated leachate collected from the Welcome WMF. In addition, pilot testing provided operating data required for the full-scale water treatment plant process design [12].

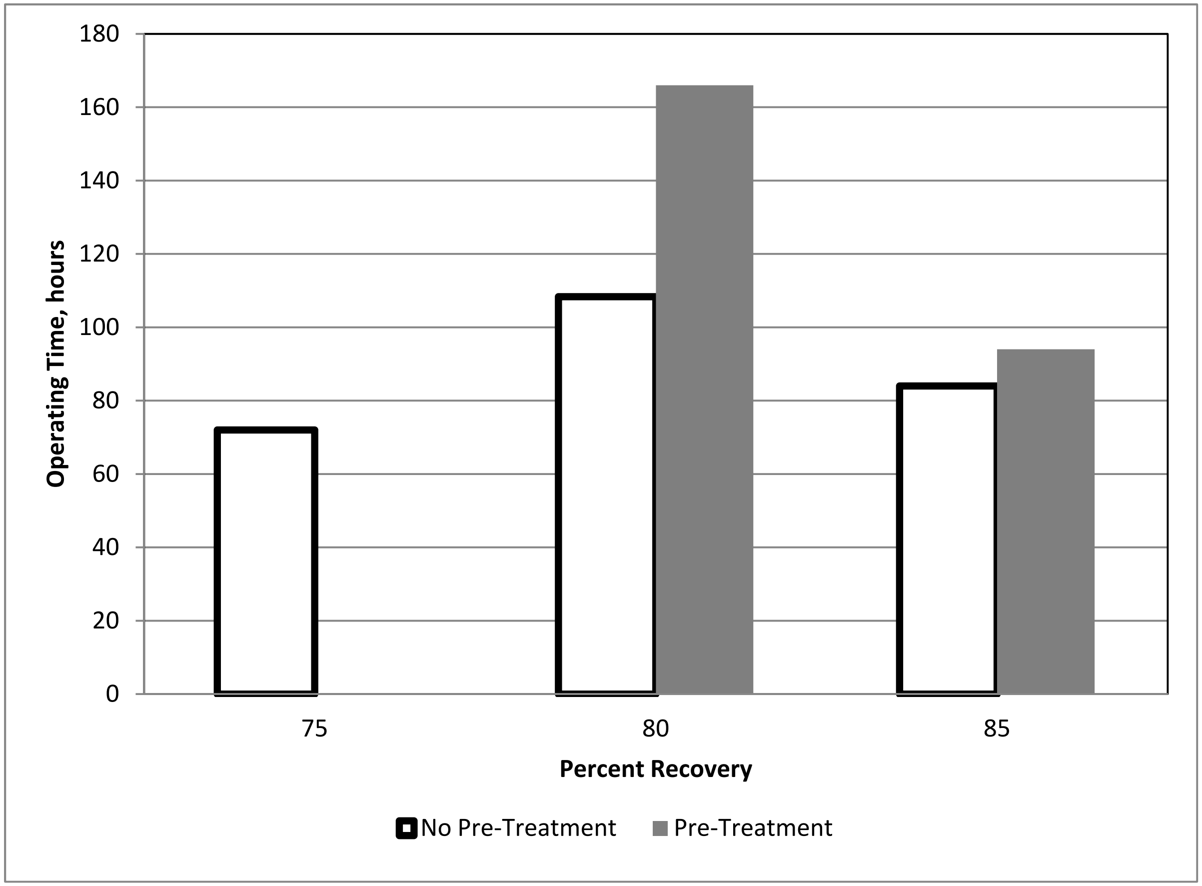

Fig. 4: Bar graph of operating time versus percent recovery with no TDS spike. The percent recovery refers to the percent of permeate flow with respect to influent flow into the first reverse osmosis (RO) module. Pretreatment refers to the addition of 130 ppm $FeCl_2$ flocculent in the clarifier whereas no pre-treatment refers to no flocculent addition in clarifier. The conductivity of the influent to the first RO module was not altered; referred to as no TDS spike. The operating time refers to the time between chemical cleaning cycles for the RO pilot system. The cleaning cycle is activated when the applied membrane pressure increases above 30% of the starting applied membrane pressure.

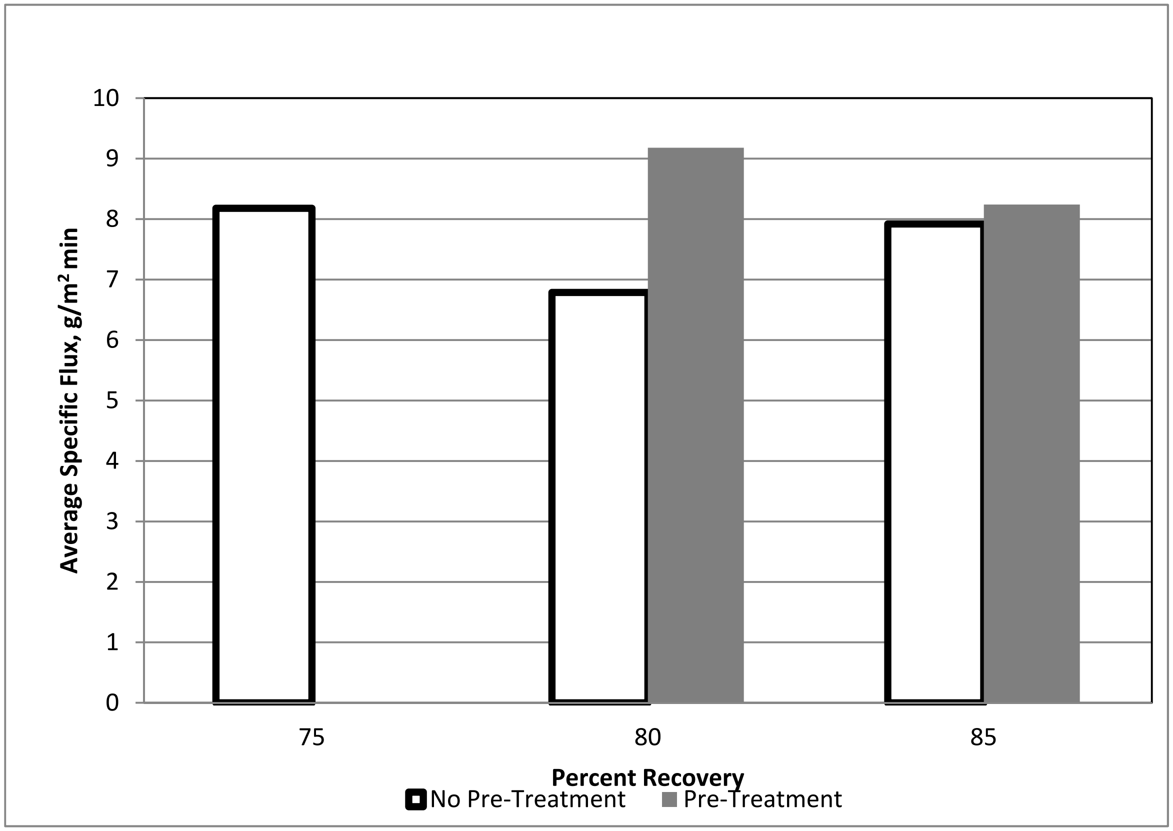

Fig. 5: Bar graph of average specific flux versus percent recovery with no TDS spike. The percent recovery refers to the percent of permeate flow with respect to influent flow into the first reverse osmosis (RO) module. Pretreatment refers to the addition of 130 ppm $FeCl_2$ flocculent in the clarifier whereas no pre-treatment refers to no flocculent addition in clarifier. The conductivity of the influent to the first RO module was not altered; referred to as no TDS spike. The average specific flux refers to the flux normalized with respect to temperature and applied pressure.

Table III
Percent Rejection of Contaminants of Concern for
Statistical Experimental Design[i] Results with No TDS Spike[ii]

Pre-Treatment/ No Pre-Treatment[iii]	Percent Recovery[iv]	Percent Rejection		
		Uranium[v]	Radium-226[vi]	Arsenic[vii]
No Pre-Treatment	75	99.7	90.0	99.7
	80	99.6	91.7	99.6
	85	99.6	50.0	99.6
Pre-Treatment	80	98.9	75.0	99.7
	85	99.6	83.3	99.8

i Design of Experiment (SDOEX): CRA-LTD evaluated three main effects with first order interactions and higher considered as error. The SDOEX was a crossed three-way mixed unbalanced model design to evaluate data from nine runs with three repeats.

ii TDS Spike refers to injecting collected concentrate from the RO unit previous runs into the influent of the first RO module. Concentrate was added to increase the electrical conductivity in the range of 1,000 to 1,200 uS/cm. No TDS Spike means the electrical conductivity was not altered.

iii Pre-treatment refers to ferric chloride dosage in the clarifier upstream of the RO unit. Levels evaluated were pre-treatment (ferric chloride dosage at 130 ppm) and no pre-treatment (no ferric chloride added).

iv Percent Recovery refers to percent of permeate flow to that of the influent flow to the RO system on a volume basis.

v Uranium Rejection refers to the percent of uranium (mass) retained in the concentrate from the RO module to that in the feed of the RO module.

vi Radium-226 Rejection refers to the percent of radium-226 (radioactivity in becquerel) retained in the concentrate from the RO module to that in the feed of the RO module.

vii Arsenic Rejection refers to the percent of arsenic retained (mass) in the concentrate from the RO module to that in the feed of the RO module.

CONCLUSIONS

The on-site pilot study performed by Conestoga Rovers & Associates (CRA-LTD) evaluated the proposed CRA-LTD leachate treatment process design for the removal of low-level radioactive contaminates as well as provided effluent water that met the surface water discharge criteria mandated by the Canadian Nuclear Safety Commission. The design of experiment applied evaluated the main effects of factors: percent of water recovery, coagulant dosage, and spike in total dissolved solids on the measured responses: operating time, average specific flux, and rejection of radioactive contaminants along with other elements. The analysis of variance (ANOVA) results for the data collected from the on-site study revealed that the maximum operating time can be achieved when an 80% recovery is applied and the addition of 130 ppm of $FeCl_2$ as a flocculent in the clarifier. This operating time was observed to be more than 50% greater than the operating time for the other conditions evaluated. In addressing the operating conditions of the treatment process that maximizes the operating time the minimum overall operating costs for the process should be achieved.

The ANOVA results revealed the rejection for the contaminants was unaffected by the levels of the factors evaluated. Based on this observation it is conjectured that the final effluent water quality generated by the treatment process may not be significantly affected if the system strays from the set points for the different unit operations. This is based on the operating range of the data collected in the study. The CRA-LTD leachate treatment process design was observed to be robust process. This is based on 3 months of continuous on-site testing of the pilot system of the impacted leachate without process issues. The required data for sizing the full scale leachate treatment plant was obtained from this study.

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