Selective removal of plutonium 238 from a canal sediment using a carbonate-chelant soil washing technology (ACT*DE*CON^{SM}) [1]

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Abstract

The Mound laboratory site in Miamisburg, OH, a former plutonium processing facility, contains approximately $40\,000$ yd^3 (30,580 m^3) of plutonium- and thorium-contaminated soils and sediments at levels that require remediation. Existing applicable remediation technologies are unsatisfactory, because they are expensive and do not provide volume reduction. ACT*DE*CON^{SM} is a chemical soil leaching technology for the treatment of soils that utilizes contaminant dissolution via dilute selective solutions to remove radionuclides. In bench-scale tests, process parameters were developed for the optimal treatment of the Miami Erie Canal soil at the Mound site, combining the maximum plutonium removal with an acceptable amount of soil dissolution and minimizing the costs of reagents. Parameters evaluated included soil to extractant mass ratio, temperature, rinse solution composition, kinetics, and the application of several dewatering aids. Plutonium removal rates of $> 95\%$ were achieved, and the residual plutonium in the treated soil proved to be very immobile—confirming that the process had removed the most accessible species of the radionuclide. Currently being tested at Mound is an engineering scale-up that includes an attrition scrubber, a counter-current extractor, and a reverse osmosis system.

Economic evaluations based on bench-scale results put the treatment cost at US\$278/yd^3 (US\$364/m^3), compared to US\$350/yd^3 (US\$458/m^3) for the 'box-and-bury' baseline alternative treatment system. © 1999 Published by Elsevier Science B.V.

1. Introduction and background

The U.S. Department of Energy (DOE) and the U.S. Environmental Protection Agency (EPA) have identified contaminated sediment as a major problem at several U.S. Government sites. The Mound laboratory, a former plutonium processing facility in Miamisburg, OH, and the close-by Miami Erie Canal, are among these. Part of the abandoned Miami Erie Canal, paralleling the Great Miami River, receives part of the runoff and the storm water discharges from the Mound Laboratory. In 1969, a low-level plutonium leak contaminated 1.5 miles (2.4 km) of soil and sediment within the old canal system. An estimated 20 000 yd^3 (15,300 m^3) of soil/sediment in the canal requires remediation. The canal overlies the Buried Valley Aquifer, which is 20-ft (6.1 m) below. An estimated 5 Ci of plutonium 238 (^{238}Pu) (18.5 × 10^{10} Bq (becquerel)) is in the canal sediment [15]. Expected regulatory limits for the cleanup are 75-pCi/g soil (2.8 Bq/g) at the 95% upper confidence limit and 150 pCi/g (5.5 Bq/g) maximum.

The soil/sediment from the Miami Erie Canal near the Mound site has a high silt and clay content. Approximately 80% of the sediment particles are less than 32 μm in diameter, with 33% less than 2-μm [4]. Most of the plutonium is associated with the smaller soil particles. Conventional soil washing, which uses particle size separation techniques, was rejected as a treatment option because it would not reduce the volume of waste enough to be economically acceptable. Furthermore, the Mound site operators would prefer reusing the treated soil as a fill material, returning it to the locations from which it was originally removed. Therefore, minimizing soil loss through selective leaching and achieving a final soil quality compatible with this reuse are important. Critical components of a viable remediation technique for the cleanup of the Miami Erie Canal soil are (1) that soil loss during the treatment be controlled, (2) that the treated soil not contain any of the readily mobile Pu, (3) that the soil's mechanical characteristics be compatible with its reuse, and (4) that the soil can sustain vegetation to minimize wind erosion and runoff.

Selective Environmental Technologies (Selentec) has developed the ACT*DE*CON^{SM} process for use in soil and sediment to chemically dissolve and remove metal and radioactive contaminants. The process combines contaminant dissolution, via dilute selective solutions, with contaminant and solution recovery to provide a continuous, recirculating treatment to remove radionuclides and heavy metals from soils and sediments. A variety of chemical concentration ratios can be used, depending on the target contaminant. The solution chemistry used to remove this actinide combines established carbonate recovery chemistry with a chelant in an oxidative environment. The oxidant is required to raise the oxidation state of the contaminant to allow formation of plutonium carbonate complexes. The function of the chelant is not fully understood, but it is observed to be an important factor in the formation of soluble complexes.

A speciation study of the soil guided the formulation of the washing solution to address the species of plutonium most commonly represented in the Miami Erie Canal soil, notably the ones associated with plutonium oxides and the soil's organic matter [1]. Once a formulation and solids loading was demonstrated as effective in preliminary studies [1], a bench-scale testing of the process parametric conditions was performed, that provided the basis for further engineering of the process and economic evaluations. The chosen testing conditions for the bench scale were the ones that had previously been proven to be usable for scale-up.

2. Objective

The objective of this paper is to discuss the results of research performed to optimize the $ACT^*DE^*CON^{SM}$ process to cleanup the Miami Erie Canal soil.

In particular, this paper will discuss:

· Optimization of the process parameters to develop an operating envelope for pilot-scale application and eventual full-scale application,
· Effects of the treatment on the dissolution of soil and of nonhazardous elements present in the soil,
· Mobility of the residual plutonium in the treated material (K_d studies), and
· Economic and engineering aspects of the full-scale application of the technology for the remediation of the Miami Erie Canal soil.

A detailed study of the conditioning and revegetation of the treated soil is currently under study and will be the subject of future publications.

3. Bench-scale experiments

3.1. Materials and methods

3.1.1. Soil description

The contaminated Miami Erie Canal soil under study consists of a mixture of deposited soil and sediment (for practical purposes, it is referred to here as soil) from the currently abandoned Miami Erie Canal, which lies a short distance away from the Mound laboratory's western fence line. Part of the abandoned canal paralleling the Greater Miami River receives the runoff and storm water discharges from the Mound laboratory. In 1969, a low-level plutonium leak contaminated soil and sediment 1.5 miles (2.4 km) downstream, in the old canal system. An estimated 5 Ci of ^{238}Pu (18.5×10^{10} Bq) is in the canal along this stretch.

The soil indigenous to the Mound site is a glacial till, with Miamian silt, clay loam, and Fairmont silty clay loam being the predominant soils. Several particle size distribution analyses were performed that indicated quite consistently that clays ($< 2 \mu m$) make up about 30% of the soil in the Miami Erie Canal, while another 52–56% is silt and 10–15% is sand [1–3]. Clays in the canal soil are mica (illite), montmorillonite, and other expandable clays, vermiculite and kaolinite. Additional components of this soil are

carbonates (approximately 20%), namely calcite and dolomite, limestone, and organic matter [4,5]. Typically, this soil has a pH of 8, contains 3% organic carbon content, and has a cationic exchange capacity (CEC) of about 35 cmol/kg.

The average plutonium activity levels in the canal soil are around 500 to 600 pCi/g (18 to 22 Bq/g) with 'hot spots' reported at up to 4600 pCi/g (170 Bq/g). Analyses conducted by the Mound laboratory in the past [2] indicated that about 69% of the plutonium activity were associated with the clay fraction, and a lower amount (24%) with soil particles between 5–20 μm in diameter. Only 6% of the activity were associated with soil particles larger than 20 μm in diameter.

In previous studies [1], the sediment was also examined to evaluate plutonium associations in the sediment by means of a sequential extraction procedure. The analysis showed that the majority of the plutonium in the soil was either complexed with organic material in the sediment (52%) or associated with oxides (41%). A small fraction of the plutonium (3%) was classified as residual. This fraction, analyzed by total dissolution of the sediment, was considered to be in a form that is either fused with the sediment or chemically complexed with the sediment particles. A very small fraction of the plutonium (0.2%) was readily removed from the sediment by a bland extractant such as water or by a salt extraction.

At the start of the bench-scale test, soil previously removed from the canal at a 'hot spot' was placed in a large Hobart planetary mixer and blended to achieve homogenization. Analysis of duplicate grab samples showed that the average ^{238}Pu concentration was 2545 pCi/g (94 Bq/g), that is believed to be much higher than the average contaminant level in the canal sediment. Analysis of the metals content of the soil reported 7.2% by weight calcium, 1.4% iron, 0.2% potassium, 2.4% magnesium, and 0.03% manganese. The bulk density was 1.2 g/cm^3, and the moisture content of the blended material was 21.3%.

3.1.2. Optimization of bench-scale process parameters

A testing protocol was planned in a cascade style, where one parameter at a time was studied at various levels; the best of these levels was then included as a test condition into the testing of the subsequent variable. All other conditions were kept constant. Fig. 1 shows a schematic of the test plan.

Standard conditions adopted throughout the tests included the basic formulation of the ACT*DE*CON^{SM} solution as optimized in previous phases of work, which included previously standardized concentrations of disodium ethylenediaminetetraacetic acid (EDTA), sodium bicarbonate, and hydrogen peroxide, and an alkaline pH (obtained by adjustment with sodium hydroxide) [1]. The 'standard' extraction consisted of two consecutive extraction stages (2 h each), plus a single rinse. The 'standard' batch size was 2000 ± 200 ml of slurry. Standard test temperature was 50°C. All tests were run in duplicate.

The tests were carried out using a Denver flotation cell [6] with an attrition impeller (two opposing-pitched propeller blades) as the agitation-mixing device in a square reactor vessel. This high-shear mixing unit ensured that the extractant solutions made thorough contact with the contaminated soil. This test scenario had been demonstrated in other programs [7] to provide results usable for scale-up in an attrition scrubber,

1. SOIL SOLIDS LOADING

Variables: 5, 10, 15%
solids liquid ratio.

Fixed Conditions:
2 extractions of 2 hr each
at 50°C.
1 rinse w/DIW (pH 7).
No dewatering additives.

2. RINSE SOLUTION

Variables: Deionized water
at pH 7 and pH 9; 0.1 M
KNO$_3$, pH 9 all at 50°C.

Fixed Conditions:
2 extractions of 2 hrs each,
at 50°C, 10% solids.
No dewatering additives.

4. DEWATERING ADDITIVES

Variables:
Haborlite —- 200
Catfloc L
Percol 351

Fixed Conditions:
10% solids
Rinse with DIW pH 9,
2 extractions, 2 hr, 70°C

3. TEMPERATURE

Variables: 50° and 70°C.

Fixed Conditions:
2 extractions of 2 hrs.
each, 10% solids.
1 rinse with DIW pH 9.
No dewatering additives.

5. KINETICS TEST

Variables: Sampling
of liquid at 1, 2, 3, 4, 5, and
6 hrs contact, and after rinse.
Sampling of solids at 2 and
6 hrs contact, after rinse.

Fixed Conditions:
2 extractions, 1st of 2 hrs
and 2nd of 4 hrs.
10% solids, rinse with DIW pH 9.
70°C for extraction and rinse.
No dewatering additives.

6. SEQUENCED FULL TEST

Variables: Soil at 2 activity
levels, average soil with and without
dewatering additive extraction.

Fixed Conditions:
High Pu soil: 10% solids.
5 extraction stages of 1 hr each.
70°C average Pu soil: 3, 1 hr
extraction stages, 10% solids.
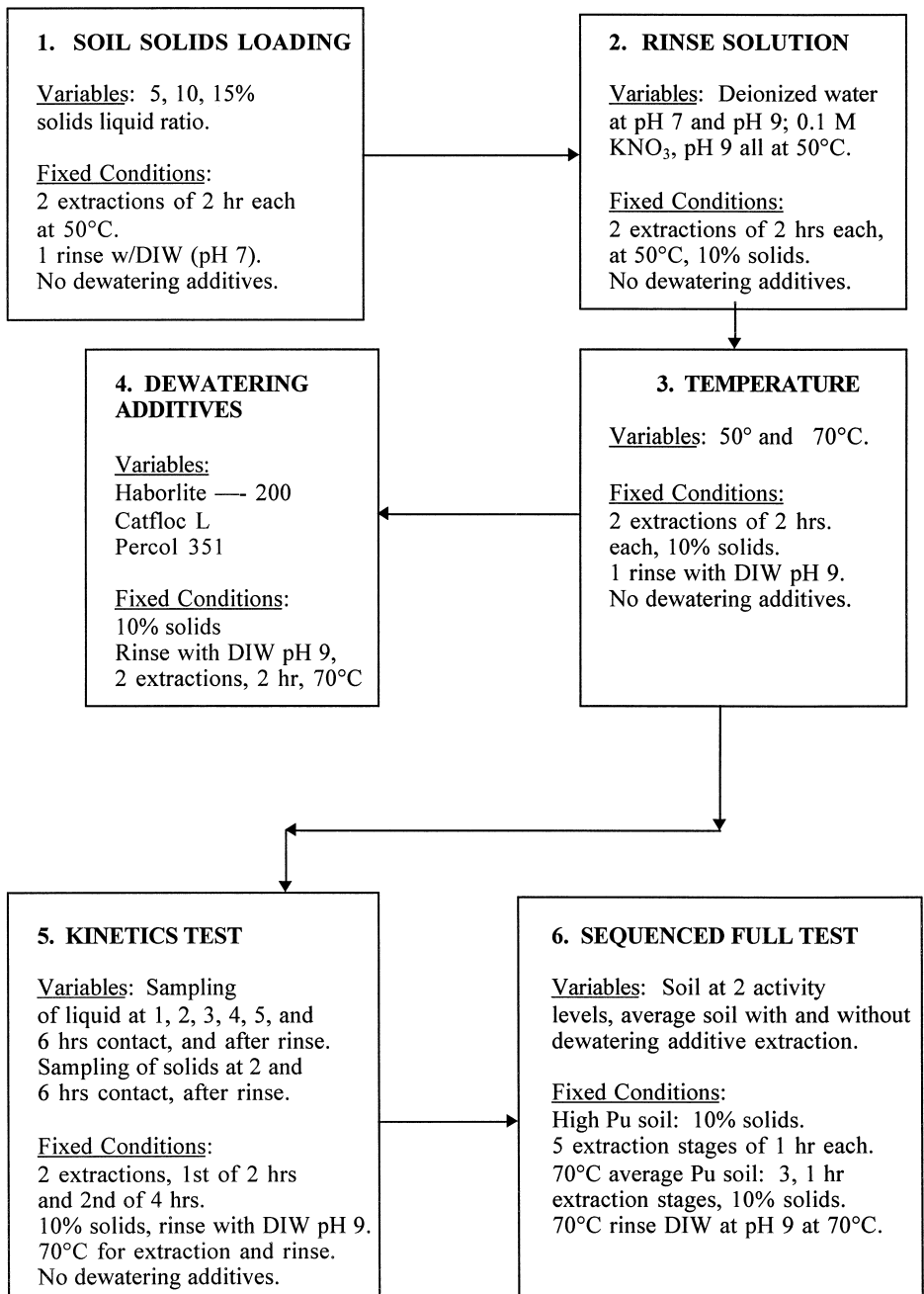70°C rinse DIW at pH 9 at 70°C.

Fig. 1. Bench-scale process optimization flow diagram.

followed by a multiple-stage, batch extraction system. After the required contact time, the slurry was vacuum filtered with 0.7–1-μm filter paper. The cake solids were then reslurried with fresh $ACT^*DE^*CON^{SM}$ (second stage) and, at the end, with deionized water and vacuum filtered again to rinse out most of the reagents. All the fractions were analyzed separately. To account for differences due to possible soil inhomogeneities, the initial total soil concentration used for determining Decontamination Factors (DF) and removal rates was calculated for each sample by quantitatively adding all of the Pu in the different fractions. The bench-scale investigation included monitoring Pu removal rates, percent soil dissolution, removal of nonhazardous elements, and EDTA utilization under the following process variables: (1) *Solids loading*: concentrations of 5, 10, and 15% soil solids in $ACT^*DE^*CON^{SM}$ were compared. (2) *Rinse solution*: deionized water at pH 9 and pH 11 (both adjusted with KOH) was compared with a 0.1-M solution of KNO_3 adjusted to pH 9 (always with KOH). (3) *Elevated temperature*: an operating temperature of 70°C was compared with the standard one of 50°C. (4) *Kinetics test*: the effect of time was investigated to evaluate the advantages of extended solution contact. (5) *Dewatering additive*: previous studies on soil handling recommended the use of dewatering aids to achieve appreciable solid–liquid separation. The best-performing additives were tested to understand their potential interferences with Pu mobilization.

To evaluate the levels achievable from a full application, a sequenced extraction was then conducted, incorporating all of the best-performing conditions from all of the previous tests in three replications, multiple-stage extraction, and rinse tests. This procedure was expected to closely model the effects of a counter-current extractor (CCE) (Fig. 2), the selected method of application at the pilot and full scales. The procedure was then repeated on a sample of canal sediment at plutonium contamination levels closer to the typical activity of 500 pCi/g (18 Bq/g), to confirm the effectiveness of the technology under more realistic conditions.

Analytical methods used were as follows: pH, EPA method 9045; isotopic Pu by α Spectroscopy by CTC method 92-69; minerals (Ca, Fe, Mg, Mn, and K), EPA method 6010A; and total/unchelated EDTA, ASTM method D3113-87. All aqueous samples were digested according to EPA method 3010A, and all soil/solid samples were digested according to EPA method 3050A.

3.1.3. Plutonium mobility tests

The objective of these studies was the investigation of the mobility of the residual Pu not removed by the $ACT^*DE^*CON^{SM}$ treatment, as an indication of its potential for being taken up by vegetation and/or to be leached through the sediment profile into the aquifer. For this investigation, batch-mode distribution coefficient (K_d) studies were used in a comparative test between treated and untreated soil. K_d is defined as the ratio of the concentration of the element in the soil (dry basis) to the concentration of the element in the aqueous phase that is in equilibrium with the soil (expressed in milliliters per gram [ml/g]).

Data from previous work [1] suggested a need to supply the treated soil with amendments in relatively large amounts to restore soil for reuse. Addition of organic matter has been related (although this remains controversial) both to increased immobilization of Pu and to its potential solubilization by chelation by soluble fractions of
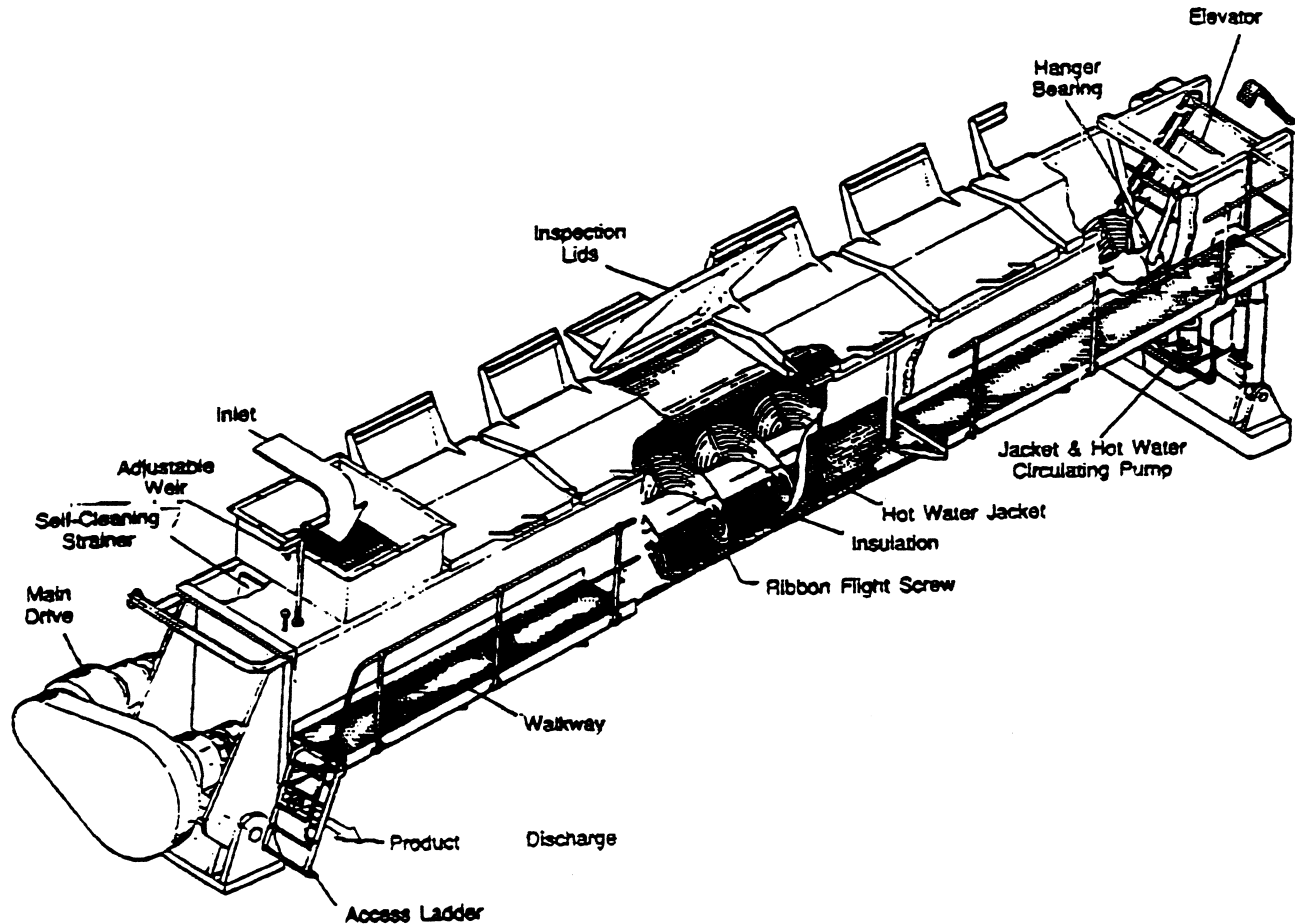
Fig. 2. Illustrates the counter-current extractor (CCE). The counter-current extractor (CCE) technology is owned by Processing Technologies International, and was developed for the food industry. The CCE technology is licensed to PWS Technology for use in heavy industrial process applications developed by PWS.

decaying organic matter. Therefore, the effect of amendments on Pu mobility was also investigated in this study.

Contaminated, moist soil sediment obtained from the Miami Erie Canal was thoroughly mixed in a plastic tray, cleared of evident stones and root debris, and sampled for Pu activity and moisture analysis. Nine aliquots of moist soil, each equivalent to 40 g of dry soil, were accurately weighed and placed in nine plastic, 1000-ml centrifuge bottles. Three bottles were sealed and set aside as controls, and the other six bottles of soil were subjected to $ACT^*DE^*CON^{SM}$ treatment according to standard procedures. Initial and final ^{238}Pu activities in this soil were 844 and 77.3 pCi/g, respectively, on a dry-weight basis.

Three days after completion of the $ACT^*DE^*CON^{SM}$ treatment, the treated soil in three of the bottles was individually slurried with 80 ml of deionized water, neutralized with 0.1 N sulphuric acid to a pH of 8–8.5, and then centrifuged for 16 min to remove the excess liquid (pH of the supernatant liquid was 8.35). To each bottle, 3 g of dry yard-waste compost, 10 mg of KH_2PO_4, 26 mg of urea, and 0.5 ml of mixed, nonchelated microelements were added to satisfy requirements of fertilizer amendments. After thorough mixing, rechecking, and adjusting the soil moisture to the same level, all nine bottles were placed in the incubator at 16–18°C.

A composite sample of the treated soil was prepared for the first K_d contact at day four after soil washing, by retrieving 1 g of material from each of the treated bottles and mixing them together with a glass rod. A 1-g aliquot of the mixture was dried at 105°C to determine the moisture content and was analyzed for Pu. Two other aliquots of the mixture were weighed to obtain the equivalent of 0.300 g of dry soil, and each was placed in a 500-ml glass flask. In a similar manner, a cumulative sample of the three control soil samples was prepared, and equivalent aliquots were also weighed and placed in flasks. To each flask, 300 ml of a sample of rainwater collected at the Mound site was added; the flasks were then sealed with parafilm, and their contents were gently stirred for 6 days. After 6 days, an aliquot of the soil suspension was filtered with a 0.45-μm millipore membrane and sampled for Pu analysis.

At 19, 30, and 90 days from the beginning of the experiment, an aliquot of soil from each bottle was weighed (0.300 g of dry soil) and contacted for 6 days with 300 ml of rainwater, according to the previously described method, to determine the second, third, and fourth K_d value series.

3.2. Results and discussion

3.2.1. Bench-scale testing

A summary of the results from the bench-scale tests is provided in Tables 1–10.

Plutonium removal from the 5% solids loading was clearly higher than the removal obtained at higher loadings. This was expected, as a consequence of the greater availability of reagents per mass of soil, and was also supported by the data on the chelation of the EDTA during the process. At the end of the first stage, about 27% of the EDTA was in the unchelated form in the 5% test, dropping to 1.6–0.1%, respectively, in the 10 and 15% loadings. At the end of the second extraction stage, while the 15% test was still clearly overloaded, with only 2.8% EDTA unchelated, the 5% solids showed a

Table 1
Plutonium removal under different process conditions

Treatment	Pu removal (%)	DF[a]	Residual Pu (dry wt. basis)[b]	Calculated starting activity (dry wt. basis)[b]
5% solids (1)[c]	92.2	12.7	223 (8.2)	2160 (80)
10% solids (1)[c]	89.2	9.3	346 (12.8)	2471 (91.5)
15% solids (1)[c]	85.2	6.8	374 (13.8)	2114 (78.3)
DIW pH 9 (2)[c]	92	12.4	214 (7.9)	n/a
DIW pH 11 (2)[c]	90.6	10.7	253 (9.4)	n/a
0.1 M KNO$_3$ pH 9 (2)[c]	90.6	10.6	257 (9.5)	n/a
70°C (3)[c]	93.1	14.4	191 (7.1)	2139 (79.2)

[a] DF: Pu in original soil divided by Pu in treated soil.
[b] pCi/g (Bq/g).
[c] Numbers in parenthesis refer to test number of Fig. 1.

clear underloading of the EDTA, which was 89% unchelated. The 10% solids showed a more balanced 35% of unchelated EDTA.

Testing of the rinse solutions was performed at 50°C, to evaluate the effect of different rinse solutions on the overall removal process. The three treatments yielded very similar results for Pu removal; all were slightly better than the 'standard' rinse, which included deionized water at pH 7. In addition, these rinses also reported a decrease in the amounts of soil lost in the treatment. The idea behind the utilization of alkaline pH and saline rinses was to facilitate Pu removal while achieving a substitution of sodium, which saturates the clay's exchange complex at the end of the treatment, with potassium. Potassium may be considered a preferable cation on the treated soil due to its positive effects on clay reflocculation and soil fertilization. Further study on this subject is under way. The reduced amount of soil lost by rinsing the clays at higher pH or with a saline rinse was considered a very good indication that the hard-to-settle fines of the treated soil were more easily handled and that less of them were lost during the filtration. Among the three rinses for subsequent application, the deionized water rinse at pH 9 was chosen not only because of its tendency to outperform the other two, but also in view of the lesser costs for reagents needed.

Table 2
Removal of soil and nonhazardous elements (percent of initial content lost)

Treatment	Soil solids	Ca	Fe	K	Mg	Mn
5% solids (1)[a]	24.0	52.7	11.9	33.8	5.2	59.8
10% solids (1)[a]	23.0	55.3	11.9	25.8	6.0	60.0
15% solids (1)[a]	16.3	41.1	7.6	22.8	1.7	51.1
DIW pH 9 (2)[a]	17.1	53.9	11.0	28.9	4.6	57.8
DIW pH 11 (2)[a]	17.1	54.5	9.4	29.8	4.7	54.3
0.1 M KNO$_3$ pH 9 (2)[a]	17.1	52.3	9.2	n.a.	4.3	51.6
70°C, 10% solids (3)[a]	22.2	53.5	7.2	27.7	5.3	45.5

[a] Numbers in parenthesis refer to test number in Fig. 1.

Table 3
EDTA utilization results

Treatment	Unchelated/total EDTA (%)	
	End of first stage	End of second stage
5% solids, 50°C (1)[a]	27.2	89
10% solids, 50°C (1)[a]	1.6	35.4
15% solids, 50°C (1)[a]	0.1	2.8
70°C, 10% solids (3)[a]	5.9	40.1

[a] Numbers in parenthesis refer to test number in Fig. 1.

Elevated temperatures of 70°C raised the amounts of Pu removed from the soil at 10% soil loading, compared with the standard condition of 50°C. At 70°C, a Decontamination Factor (DF) of 14.4 was obtained, equivalent to a 93.1% removal of Pu. Tests conducted at 70°C confirmed the data obtained at 50°C with regard to EDTA utilization and showed that greater Pu removal was achieved with more efficient use of the EDTA at higher temperatures.

With respect to the dissolution of nonhazardous elements, between 40–54% of the soil calcium, more than 50% of Mn, approximately 30% of potassium, and less than 10% of iron and magnesium were lost with the treatment. This converts, based on the original soil concentration, to approximately 3.24% Ca, 0.015% Mn, 0.06% K, 0.14% K, and 0.24% Mg. Total nonhazardous minerals lost were on the order of 3–4% of the total initially present in the soil. The lowest values were achieved at 15% solids loading, and the highest, with the pH 11 rinse.

To minimize soil loss and achieve faster cake dewatering, trials were conducted where the slurry from the rinse step was added with one of three filter aids (previously selected during a screening phase). As seen in Table 4, the two polymeric filter aids did not seem to interfere significantly with the removal of Pu. They generated a filter cake that was wetter than the no-additive control, but they significantly accelerated the filtration times. The other compound, a diatomaceous earth frequently used to dewater fines, was also efficient at reducing filtration times, but it appeared to retain significant

Table 4
Pu and soil balance with use of dewatering aids

Treatment	Pu removal (%)	DF	Final/ initial soil mass (%)	Additive/ soil mass (%)	Solids in cake[a] (%)	Filtration time (min)
No additives	85.9	7.1	66	0	98.7	> 900
Diatomaceous earth	72.9	3.7	175	66.7	48	1.5
Cationic polymer/ surfactant	84.3	6.4	98	2.3	82.7	0.3
Polymer/surfactant	83.6	6.1	116	0.1	59.7	30.0

[a] Includes solids from extractant solution.

Table 5
Kinetics test results

Hours from start	Pu activity, pCi/g (Bq/g)	Initial Pu (%)	Initial soil recovered (%)	Unchelated/ total EDTA (%)
Initial (calculated)	2182 (80.8)	100	100	100
1 h	455 (16.8)	20.8	n.a.	1.7
2 h	502 (18.6)	23.0	90.5	2
3 h	180 (6.7)	8.2	n.a.	43.3
4 h	190 (7.0)	8.7	n.a.	32.5
6 h	167 (6.2)	7.6	n.a.	35.4
After rinse	57.4 (2.1)	3.3	83.3	n.a.
Overall Pu removed	2125 (78.7)	97.2	–	–

amounts of water, radioactivity, and process chemicals, as shown by the increase of mass at the end of the test and by the solids content.

While precise information on the minimization of soil loss by using filter aids was not obtained, the results represented the first step in finding a suitable dewatering agent to help minimize both soil loss and the time needed to dewater the slurry.

The kinetics tests (in which a 6-h test was conducted, with fresh extractant added after the first 2 h of contact) indicated (Table 5) that the overwhelming majority of the Pu activity was removed within the first hour of contact, in both the first and second stages. Soil loss also appeared to be proportional to the number of stages and filtrations involved. The rinse with deionized water at pH 9 appeared to have a dramatic effect on the removal of Pu from the solids; the cake activity went from 167 before to 57.5 pCi/g (6.2–21 Bq/g) after the rinse. The primary result from the kinetics test that was utilized in the design of the ensuing work was that, to maximize process efficiency, the contact time for each extraction stage should be limited to 1 h.

Table 6
Sequenced extraction test results[d]

Test conditions	Initial and final activity, pCi/g (Bq/g)	Pu removal (%)	DF	Unchelated/ total EDTA (%)	Soil recovered (%)
70°C[a]	2179–79 (80.7–2.9)	97.2	35.6	57.7	77.5
70°C[b]	803–66 (29.7–2.4)	92.3	12.2	n.a.	93.9
70°C +[c] filter aid	793–62 (29.4–2.3)	90.2	10.1	n.a.	94.1

[a] Five extraction stages, plus two rinses.
[b] Three extraction stages, one rinse.
[c] Same as b, but with filter aid.
[d] Fixed conditions: 10% solids loading, rinse with deionized water at pH 9 at 70°C, 1 h each extraction stages.

Table 7
Plutonium extraction in the four K_d contacts

Days	Plutonium extracted, fCi/L (Bq/l)		
	Treated soil	Treated/fertilized soil	Untreated soil
4	137[a] $[5.1\times10^{-3}]$	n/a[b]	1879[a] $[7\times10^{-2}]$
	(4)[c] $[1.5\times10^{-4}]$		(1197) $[4\times10^{-2}]$
19	112[d] $[4.1\times10^{-3}]$	292[d] $[1.1\times10^{-2}]$	3964[d] $[1.5\times10^{-1}]$
	(51) $[1.9\times10^{-3}]$	(26) $[9.6\times10^{-4}]$	(1369) $[5\times10^{-2}]$
30	325[d] $[1.2\times10^{-2}]$	856[d] $[3.2\times10^{-2}]$	11,193[d] $[4.2\times10^{-1}]$
	(145) $[5.4\times10^{-3}]$	(140) $[5.2\times10^{-3}]$	(678) $[2\times10^{-2}]$
90	514[d] $[1.9\times10^{-2}]$	488[d] $[1.8\times10^{-2}]$	5891[d] $[2.2\times10^{-1}]$
	(237) $[8.8\times10^{-3}]$	(187) $[6.9\times10^{-3}]$	(704) $[3\times10^{-2}]$

[a] Mean value of two replications.
[b] n/a—not applicable.
[c] Numbers in brackets give the standard deviation.
[d] Mean value of three replications.

Sequenced extraction tests (Table 6) were designed to incorporate all of the positive impacts from the preceding tests into a 'process' that would have the best likelihood of achieving the lowest residual Pu in the treated soil. These extractions were conducted on the high activity soil and on one soil sample with a plutonium activity level of 800 pCi/g (30 Bq/g), which is more representative of the actual, average contamination levels. Pu removal of $> 90\%$ was achieved in all cases, with a maximum of 97% in the high-Pu soil. The percent removals of Pu obtained from the '3-stage wash + 1 rinse' of the lower-activity soil were slightly lower than those from the '5-stage wash + 2 rinses' of the higher-activity soil. However, the residual Pu activities in the final cake of the lower-Pu soil were comparable with those of the high-Pu soil. Further study will indicate if, and at what level, there is a threshold level of activity that cannot be removed by this treatment.

Although the Pu activities in the final filter cakes were comparable, the soil loss from the lower activity soil was clearly more contained at 6% (Table 6). It is unclear whether this decrease in soil loss is attributable solely to the lesser handling to which the lower-activity soil was subjected, however, these data represent a very acceptable rate of soil dissolution.

Table 8
Plutonium distribution in solid/liquid fractions as K_d values

Days	Treated soil	Treated/fertilized soil	Untreated soil
4	$5.7\pm1.4\times10^{5}$[a]	n/a[b]	$5.6\pm3.5\times10^{5}$[a]
19	$7.9\pm3.5\times10^{5}$[c]	$2.7\pm0.2\times10^{5}$[c]	$2.28\pm0.6\times10^{5}$[c]
30	$2.87\pm1.7\times10^{5}$[c]	$0.92\pm0.1\times10^{5}$[a]	$0.86\pm0.4\times10^{5}$[c]
90	$1.7\pm0.6\times10^{5}$[c]	$1.7\pm0.5\times10^{3}$[c]	$1.4\pm0.2\times10^{5}$[c]

[a] Mean value of two replications.
[b] n/a—not applicable.
[c] Mean value of three replications.

Table 9
Equipment design parameters for the treatment of the Miami Erie Canal sediment (field scale)

Parameter	Treatment[a]
Nominal capacity	5 yd^3 (3.8 m^3) per hour
Equipment availability	85% (estimated)
Available capacity	4.25 yd^3 (3.25 m^3) per hour
	102 yd^3 (78 m^3) per day
	3060 yd^3 (2340 m^3) per month
Processing time	9.7 months
Onsite time	10.2 months

[a] 26 000 yd^3 (19,878 m^3).

3.2.1.1. Plutonium mobility studies. At the analysis of variance (ANOVA), both K_d values and extraction values (fCi/l) obtained in the experiment showed significant differences ($\alpha = 0.01$), induced by the type of treatment and by the time of contact. Further statistical testing (Bonferoni's multiple means comparison test) showed that these differences were attributable to a number of direct comparisons between mean values of Pu dissolution and K_d.

Untreated soil gave extraction values (Pu dissolution in activity concentration values, Table 7) that were significantly different ($\alpha = 0.01$) and were approximately 10 times higher than those obtained with treated samples. Significant differences were also found between extractions from treated/fertilized and untreated soil samples at 19, 30, and 90 days. The trend toward increased extraction of plutonium in treated/fertilized soil vs. treated soil is confounded by the variability of the data and is not confirmed by the

Table 10
Application costs for the treatment of the Miami Erie Canal sediment[a]

Item	Total cleanup costs US$10^3	Cleanup costs[b] (US$)	
		Per yd^3	Per m^3
Capital equipment lease (9 months)	794	30.54	39.94
Operations and maintenance			
Spare parts (5% of capital equipment)	121	4.65	6.08
Labor (4 crews/3 shifts)	947	36.42	47.63
Utilities	423	16.27	21.28
Chemicals	1221	46.96	61.42
Consumables	372	14.31	18.72
Waste processing	1006	38.69	50.60
License fee	1053	40.50	52.97
Soil loading (operator/equipment)	180	6.92	9.05
Spreading treated soil (operator/equipment)	133	5.12	6.69
Verification analytical analysis	210	8.08	10.57
Mobilization/startup (30 days)	320	12.31	16.10
Demobilization/closeout (15 days)	133	5.12	6.69
Total to treat 20,000 yd^3	6914	267.19	349.45

[a] Estimate based on treating 20 000 yd^3 (15,300 m^3).
[b] 1 yd^3 = 0.7645 m^3.

statistical analysis. The same can be said about the dissolution from treated soil at all contact times. On the other hand, the higher degree of dissolution obtained from untreated soil at 30 and 90 days is confirmed as significantly different from the lower dissolution levels obtained during the first two contacts.

K_d values (Table 8) at 4 days from the soil treatment showed no significant difference induced by the ACT*DE*CON^{SM} treatment. At 19 days, however, K_d values obtained for treated soils were significantly higher than those obtained for treated/fertilized and untreated soils. These differences decreased with time and were not statistically significant at 90 days from the treatment. Treated and untreated soils had K_d values at 4 days that were statistically higher than those obtained at 30 and 90 days. Treated/fertilized soil had K_d values not statistically dissimilar from those of untreated soil at 19, 30, and 90 days. Differences between treated and treated/fertilized soils were significant only at 19 days.

The total amount of Pu that was leached by rainwater from the contaminated, untreated Mound soil in this experiment proved to be consistently higher (about 10 times) than the amount extracted by rainwater from the same soil after it was treated with ACT*DE*CON^{SM}. This result can be explained by the fact that treated soil was poorer in Pu, supported by the sequential extraction findings previously reported, which showed that the ACT*DE*CON^{SM} treatment removed most of the readily available and exchangeable Pu, leaving the most strongly bound Pu in the soil.

K_d values of treated and untreated soil were initially comparable and tended to decrease in both soils (i.e. to increase Pu mobilization into the liquid phase) with time. Over short intervals, the ACT*DE*CON^{SM} treatment seemed to induce a higher K_d, but this difference decreased over longer time intervals, and at 90 days the K_d values in treated and untreated soils were equal. The fertilization treatment induced a significant decrease in the K_d and made the values for treated/fertilized soil comparable with those obtained for untreated soils; in other words, fertilization increased the ratio of Pu that was extracted by the rainwater (and filtered through the 0.45-μm filter) to the level for untreated soil. At the end of the testing period, the data were equal for the three different soil conditions; thus, in terms of time interval, fertilization shortened the time needed for the residual plutonium in treated soil to reach K_d values equivalent to those of untreated soil. In no case, did the treated soil (with or without fertilization) have K_d values lower than those of untreated soils, so a higher relative mobility of the residual plutonium is not suspected.

The results obtained are consistent with those usually found in the literature for natural ecosystems. Literature data show a wide range of adsorption/desorption K_d values in soil/sediment solutions. Ranges vary between 10 and 10^6 ml/g, depending on soil characteristics, total radionuclide concentration or activity, and whether the experiment is an evaluation of environmental samples or a simulation experiment [8,9]. In the case of simulation experiments, the leaching system adopted (batch and column) and the use of artificially spiked solutions/solids are of fundamental importance in determining the K_d. Usually, the lower K_d values come from tests in which samples were artificially spiked, generally at significantly higher activity levels than those found in the samples used for this work. In nature, plutonium K_d values for marine, riverine, and lacustrine environments have been reported as rather constant at 1×10^4 to 1×10^6 [8,10,11].

Reasons for increased dissolution of radionuclides from soil (amended or not) have been connected by various authors with such factors as pH and Eh changes, the direct presence of natural organic matter (such as decaying roots, which has complexing characteristics that prevent the readsorption of Pu onto soil particles) [12,13], and the production (induced by microbes/fungi) of extracellular metabolites able to complex Pu [14]. In a study of natural waters/sediment K_d values, Ref. [11] described natural dissolved organic compounds (DOC) (such as humic materials) as important complexing agents for many metals in surface waters; they stated that the formation of water-soluble, metal–organic complexes could be responsible for a decrease in adsorption of Pu in sediments associated with waters rich in DOC.

In the present investigation, the fertilization treatment consisted of lowering the pH to a level more acceptable to vegetation (pH in fertilized soil was 8.5, compared to higher levels in unfertilized, treated soil) and in restoring some of the organic matter lost with the ACT*DE*CON^{SM} washes. Urea and potassium phosphate, as well as micronutrients, were supplied to ensure the potential for microbial life and the utilization of the organic matter supplied. Both a decrease in pH and the presence of some microbial activity might be responsible for the temporarily increased plutonium mobility.

4. Engineering design and economic evaluations

Current work is aimed at demonstrating, in an integrated pilot scale, the physical and chemical conditions at which the ACT*DE*CON^{SM} process can treat the Miami Erie Canal sediments and both thorium- and plutonium-contaminated soils at the Mound site. Pilot-scale testing is being carried out in the summer of 1997. After successful demonstration, full-scale application is expected to start in 1998. The pilot-scale testing investigates the operating conditions for a full-scale system that can process at a nominal rate of 5 yd^3 (3.8 m^3) per hour.

The principal component of the equipment design is a CCE (Fig. 2). The CCE consists of an elongated tubular trough, inclined on one end. Inside the trough is a rotor designed to produce a pulsing motion in the materials being handled. The rotor is slotted to allow for liquid flow. Solid feed enters the lower end of the unit and is propelled upward by the action of the rotor. The wash solution, injected at the higher end of the unit, passes by gravity flow through the solid material in a counter-current flow. Theoretically, a counter-current extractor has 14 extraction stages in one shell, giving high extraction efficiency while minimizing soil loss by not separating soil and liquid after each stage. The CCE has several injection points to apply the ACT*DE*CON^{SM} solution and wash water to rinse the cleaned sediment. By adjusting the inclination of the CCE, rotation speed (forward and reverse), and feed-throughput, the necessary residence times can be achieved. The CCE was preferred to a batch system because of its potential for achieving a high, sustained contact of the soil with fresh extractant, without the need for intermediate solid/liquid separations (which would involve increased soil loss and handling costs).

Fig. 3 illustrates the process flow diagram. Before entering the CEE, the soil is ground in a shredder and slurried with an attrition scrubber. The attrition scrubber is a

CHEMICAL MAKEUP

FI-6   P-4

ACT•DE•CON MAKEUP TANK T-2

SOIL HOPPER T-5

ACT•DE•CON PROCESS TANK T-3

FI-3

P-3

SHREDDER SD-1

SAND HOPPER T-1

ATTRITION SCRUBBER AS-1

HEAT EXCHANGER

TI

PI

SC-1

P-2

FI-2

FI-1

FI-5

CCE-1

P-1

CCE DRAIN TANK T-7

SAND/SOIL COLLECTION T-4

H E A T E R

BLEED

FI-7

FI-4

P-6

TI

HOLDING TANKS T-8A & B

R O SYSTEM RO-1

RECYCLE

RINSE WATER HOLDING TANK T-6

P-7   CONCENTRATE

WASTE HOLDING DRUM

EQUIPMENT LIST	
NO.	DESCRIPTION
T-1	SAND HOPPER
T-2	ACT•DE•CON MAKEUP TANK
T-3	ACT•DE•CON PROCESS TANK
T-4	SAND/SOIL COLLECTION TANK
T-5	SOIL HOPPER
T-6	RINSE WATER HOLDING TANK
T-7	CCE DRAIN TANK
T-8A	HOLDING TANK
T-8B	HOLDING TANK
CCE-1	COUNTER CURRENT EXTRACTOR
AS-1	ATTRITION SCRUBBER
SC-1	SAND SCREW CONVEYOR
SD-1	SHREDDER / DELUMPER
RO-1	RO / UF SYSTEM
P-1	RECYCLE PUMP
P-2	SOLIDS TRANSFER PUMP
P-3	ACT•DE•CON PROCESS PUMP
P-4	TRANSFER PUMP
P-5	RO PROCESS PUMP
P-6	HEATER RECIRC PUMP
P-7	DRUM TRANSFER PUMP
FI-1	RECYCLE FLOW METER
FI-2	SOLUTION FLOW METER
FI-3	SOLUTION FLOW METER
FI-4	SOLUTION BLEED FLOW METER
FI-5	SOLIDS FLOW METER
FI-6	TRANSFER FLOW METER
FI-7	HOT WATER FLOW METER
PI-1	HOT WATER SYSTEM PRESSURE
PI-2	RECYCLE PUMP PRESSURE
PI-3	SOLIDS TRANSFER PUMP PRESSURE

PROCESS FLOW DIAGRAM
ACT*DE*CON SYSTEM
MOUND PHASE IV

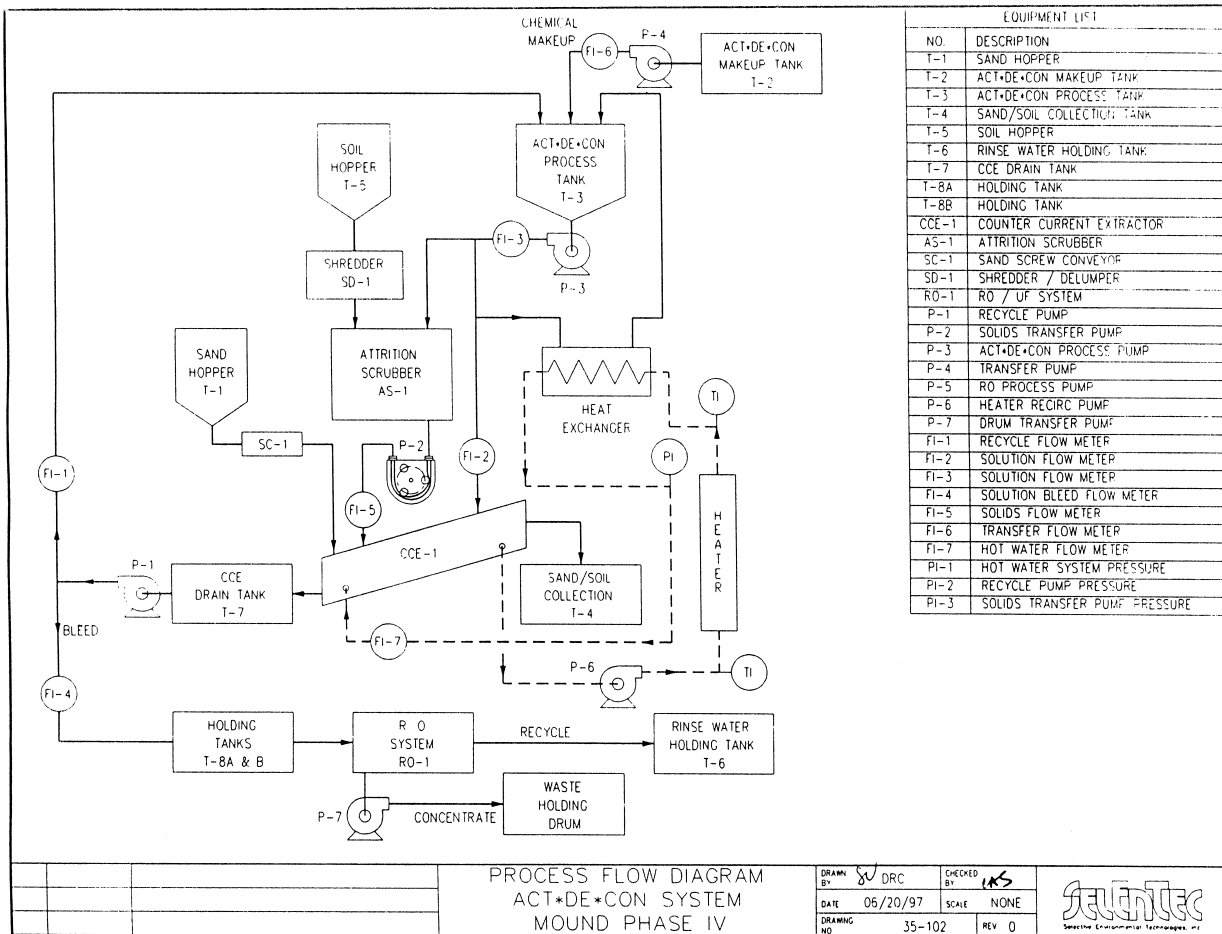| DRAWN BY	DRC	CHECKED BY	
DATE	05/20/97	SCALE	NONE
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Fig. 3. Illustrates the process flow diagram (by permission from Selentec).

scale-up of the Denver cell used in the bench-scale tests. Upon entering the CEE, the soil slurry is mixed with sand to optimize the reverse flow and contact with $ACT^*DE^*CON^{SM}$. The soil and the $ACT^*DE^*CON^{SM}$ are recovered at opposite ends of the CCE. While the soil is subject to rinsing (in the CCE) and further treatment for sand recovery, dewatering, and conditioning, a fraction of the spent $ACT^*DE^*CON^{SM}$ is pumped to a reverse-osmosis (RO) system and concentrated. The chelating agent can be recovered by acidification. The remaining concentrate is solidified and buried as LSA waste. Data on chelant recovery from the spent $ACT^*DE^*CON^{SM}$ will be included in a future publication.

The first pilot-scale test runs are evaluating the various operating parameters for the cleanup of the canal soil, including temperature, solids loading, and soil processing rates, and comparing the results with those obtained at the bench scale and reported above. Upon successful treatment of the canal soil, test runs will be performed on other plutonium- and thorium-contaminated soils from the Mound site. The information obtained will be used to evaluate the treatment of these soils with a full-scale system.

Table 9 gives the basic equipment design parameters for cleaning the canal sediment in a field application.

A preliminary economic evaluation for the field application of the technology was developed, based on the application conditions determined from the bench-scale testing and from cost and scheduling data provided by the Mound site Contractor (operator). During the operation of the pilot-scale equipment, data will be collected and used to more precisely estimate the cost of the $ACT^*DE^*CON^{SM}$ process at full scale and compare them with the costs for existing cleanup alternatives. These are currently estimated by Mound at US\$350/yd^3 (US\$458/m^3) for the baseline 'box-and-bury' technology.

Factors used to determine the processing costs include equipment operation and maintenance, personnel labor, chemicals and materials, utilities, waste processing and disposal, soil loading and treated soil spreading, mobilization and demobilization, and analytical verification.

The cost estimate is based on Mound site-specific parameters for labor costs, operating times, and soil volumes. The estimate is based on treating an estimated volume of 20 000 yd^3 (15,290 m^3) of plutonium-contaminated soil (Table 10).

Electric power consumption was calculated on the basis of known electrical ratings of treatment system components, with a nominal allowance made for auxiliaries. Steam usage rate was based on CCE field operating experience. Makeup water and sewage discharge values were calculated on the basis of known treatment system parameters. Approximate utility costs were obtained from standard engineering estimating handbooks and checked against actual utility costs, where available.

The total solvent ($ACT^*DE^*CON^{SM}$) volume was calculated using a 10:1–solvent:soil ratio and a feed and bleed rate of 5%. Solvent makeup feed will consist of water with recycled EDTA and other makeup chemicals added.

The costs of consumables were estimated on the basis of catalog and/or vendor prices for equipment required to provide reasonable personnel protection and to conduct ordinary business and administrative functions. The values for soil loading and spreading operations were provided by the Mound site Contractor and are consistent with

estimates for other treatment methods. Mound site personnel also provided the lump sum values for mobilization and demobilization, based on estimated costs for the site to provide the necessary utilities to the treatment facility.

The largest single component of the processing cost is operation and maintenance. This includes labor, materials, chemicals, utilities, and consumables, accounting for approximately 45% of the overall cost. Capital equipment costs, processing of residual wastes, and licensing fees can account for more than 40% of the treatment costs.

The capital equipment cost for the field application is estimated at US$3 707 745. The cost to the project has been estimated as a monthly lease rate of US$88 207, determined by treating the cost of the equipment as a 5-year loan at 15% interest. The lease rate would be charged for the duration of the time that the equipment is at the site, including mobilization and demobilization. The estimated cost to maintain the equipment was calculated as 5% of the monthly lease rate.

The personnel requirements for equipment operation were estimated on the basis of operating the equipment for a 24-h period, 4 days per week. Three crews were planned for three overlapping 10-h shifts, which is consistent with current practice at the Mound site. Each crew consists of two operators. Site personnel would be used as needed for support. For estimating purposes, a technician, laborer, and health physics technician were allotted to each crew, at half a person per shift for each of the three. A project manager and an administrative assistant were also factored into the cost, each with a 10-h, 4-day shift. Using the Mound working schedule, there are approximately 17 working days per month.

The total estimated cost to clean up the 20 000 yd^3 (15 290 m^3) of plutonium-contaminated Mound site soils is US$5.6 million (US$278/yd^3, or US$364/m^3) and will require 9.7 months of processing time at a nominal rate of 5 yd^3 (3.8 m^3) per hour. An equal volume of thorium-contaminated soil is also available on-site for treatment. Increasing the total volume of soil by including the thorium-contaminated soils to be treated is expected to decrease the cost to US$264/yd^3 (US$345/m^3).

5. Conclusions

Results from bench-scale testing identified the most favorable conditions for the decontamination of the Miami Erie Canal soil. These experiments highlighted the relative importance of temperature, soil solids loading, time of contact, and types of rinses and additives in the performance of the process. Effectiveness of plutonium removal was only one of the parameters that guided the selection of the most favorable conditions, the others being the amount of soil lost in the process, the efficiency of use of the reagents, economic considerations, and practical utilization in a scaled-up system.

Elevated temperature slightly increased the rate of plutonium removal, but it also increased the dissolution of the soil. Higher solid loadings of up to 15% showed decreased plutonium removal, but also a reduced soil loss rate. The use of alkaline rinses proved to maintain plutonium removal and reduce soil loss. The use of various filter aids used with commercially available filtering equipment was investigated, along with the possible interactions between filter aids and plutonium dissolution chemistry. Prelimi-

nary data showed that certain additives could decrease the time required to dewater the soil/sediment slurry by more than an order of magnitude; however, some of the additives might interfere with plutonium removal.

Sampling of the wash solution hourly, for 2 h over the first extraction stage and 4 h over the second, showed that there is very little plutonium dissolution after the first hour of each contact.

The sequenced extraction runs conducted on the canal soil at both higher and average activity levels showed that these soils, with initial activity levels of 2200 and 810 pCi/g (81 and 30 Bq/g), respectively, were remediated to levels of less than 79 and 66 pCi/g (2.9 and 2.4 Bq/g) on dry soil (66 and 55 pCi/g [2.4 and 2 Bq/g] for the sediments corrected to their natural moisture content). These levels are very close to the expected regulatory limits of 75 to 100 pCi/g (2.8 to 3.7 Bq/g) in moist soil. Soil loss in the sequenced extraction was 22.5% in the higher Pu soil, and 6% in the average Pu soil. These differences are at least in part attributable to the higher number of stages involved in the treatment of the higher Pu soil to achieve comparable residual plutonium levels.

Total plutonium dissolution and distribution coefficients were measured as functions of time and soil conditions before and after treatment. These data indicated that a larger amount of plutonium was leached from the nontreated soil, probably as a consequence of the higher content of available/exchangeable plutonium, compared to the treated soil. No increase in total relative mobility of the soil plutonium could be attributed, on the basis of these tests, to the treatment with $ACT^*DE^*CON^{SM}$ under the testing conditions. In fact, $ACT^*DE^*CON^{SM}$ treatment appeared to leave only the most insoluble forms of plutonium in the soil.

At the end of the testing, the chosen conditions for the scaled-up, pilot- and field-scale testing were a 2-stage extraction system with a rinse at 70°C, a 10% solids loading, a 1-h contact per stage, a 15-min rinse contact, and the 'standard' extraction chemistry and pH. These conditions were utilized for the design of the pilot/field-scale process equipment, in which the soil is first slurried in $ACT^*DE^*CON^{SM}$ with an attrition scrubber, then 'washed' in a counter-current extractor to achieve the required contact with fresh $ACT^*DE^*CON^{SM}$, rinsed in the same or another, sequentially connected CCE, and then conditioned and dewatered for future reuse. The spent $ACT^*DE^*CON^{SM}$ is recovered at the opposite end of the CCE, and run through a reverse-osmosis system for EDTA recovery and reuse and Pu separation.

Economic estimates (conducted using data from the bench-scale testing and available information from previous experiences with $ACT^*DE^*CON^{SM}$ and the CCE) show that this technology could be a very viable alternative to the current baseline technology of box-and-bury, reducing the treatment cost by 40%, to US$278/yd^3 (US$364/m^3), and avoiding the mere relocation of the radioactive waste to an approved landfill without improving its environmental characteristics.

The pilot-scale testing, under way in the summer of 1997, includes the processing of the Miami Erie Canal soil and other on-site Mound soil contaminated with plutonium and thorium. This pilot-scale testing will further refine the economic and technical data for the deployment of the technology and remediation of the Mound site, which has been recently selected as one of the three sites within the DOE complex designated for accelerated cleanup.

Acknowledgements

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