Pilot-Scale Study for In-situ leaching of Residual Uranium from Remediated Soil - 22204a

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ABSTRACT

A site-specific in-situ leaching (ISL) process is under development to remediate soil at a former thorium mill tailings site, known as the Former Rare Earths Facility (REF) in West Chicago, Illinois. Historical remediation activities at REF cleaned up the site soil to meet the soil cleanup standards. The remediated soil, primarily sand and gravel materials, were used to backfill three sheet-piled areas known as the Pond 1 Area, the Pond 2 Area, and the South Factory East (SFE) Area. The remediated soil contains residual levels of uranium, which continue to leach into the site groundwater at levels that exceed the associated groundwater protection standard (GWPS).

A Pilot-Scale Study was conducted to support the design of a site-specific ISL system. The objectives of the Pilot-Scale Study were to assess scale-up issues, including critical success factors and limitations, duration, cost, and the feasibility and effectiveness of the ISL technology to leach residual uranium from previously remediated soil.

The Pilot-Scale Study was conducted in an in-ground treatment cell, bounded by steel sheet piling and containing approximately 60 cubic meters of previously remediated soil. A leaching solution, comprised of 0.60 mol/L of sodium bicarbonate and 0.23 mol/L of sodium carbonate in natural groundwater, was circulated in the treatment cell to leach the uranium from soil selectively.

The pilot-scale ISL system successfully treated a large volume of soil under actual field conditions. Approximately 54% of the initial uranium mass was removed in five Pore Volume Flushes (PVFs), resulting in the attainment of the uranium GWPS. The results of the Pilot-Scale Study, consistent with previous treatability studies completed under controlled laboratory conditions, confirmed that ISL is a viable treatment option for addressing residual uranium in the REF soil. Preliminary cost estimates of the ISL alternative project a cost savings of approximately \$148 million in direct and indirect remediation costs compared to the excavation and landfill disposal remediation option.

The leachate generated during the leaching process will require treatment and disposal. Leaching of uranium resulted in unintended leaching of several non-uranium and inorganic constituents. Several of the reported constituents exceeded their corresponding site-specific effluent standards and will require management and treatment as part of the leachate treatment program.

Full-scale operations should consider alternative methods for preparing and applying leaching solutions. Accumulation of undissolved leaching solution constituents (sodium bicarbonate and sodium carbonate) on the soil surface required neutralization to halt the uranium leaching process. Groundwater acidified with HCL was successful in achieving the desired neutralization.

INTRODUCTION

The REF is spread over an area of approximately 17 hectares. Operations at the REF extracted thorium, and rare earths compounds from monazite and bastnaesite ores for use in various products. The ore extraction operations lasted from 1932 to 1973 when the plant was shut down. Waste management practices severely impacted on-site soil and groundwater, soil in neighboring residential and commercial properties,

the City of West Chicago sewer system, and about 11 kilometers (km) of the West Branch of the DuPage River and its tributary, the Kress Creek.

Remediation and investigation activities were undertaken from 1997 to 2015 to address the resulting environmental impacts. Sediments and soil were excavated and, depending on physical characteristics, disposed of either at an off-site landfill or on-site after treatment. The remediated soil met the soil cleanup criteria but continues to leach uranium at levels above the GWPS.

Initial Bench-Scale studies showed that ISL technology is a viable option to treat the REF soil. Bench-scale tests were conducted to evaluate the efficacy and viability of the ISL technology to treat previously remediated soil for attaining uranium GWPS. Specific objectives of the bench-scale tests included identifying and selecting optimal leaching parameters related to the extent and rate of uranium released as a function of chemical composition, chemical concentrations, chemical residence times, and obtaining critical data to estimate treatment costs scale-up requirements.

BACKGROUND

Site Location

The REF is located in West Chicago, Illinois, at the western edge of DuPage County, as shown in FIGURE 1. The REF is primarily surrounded by residential and commercial properties that are within the City of West Chicago limits. The land use within a 0.8-kilometer radius of the REF boundary is mixed residential, commercial, industrial, institutional, and recreational.



Fig. 1 – Site Location of the Former REF

The location of West Chicago is in the Illinois River Basin. Major drainage is towards the south and southeast. The West Branch of the DuPage River, which generally flows to the south, is east of the City of West Chicago and receives storm sewer and surface runoff from the city. Kress Creek, which is south of

the REF, flows south into the West Branch of the DuPage River. Their confluence is approximately 2.4 kms south-southeast of the center of the REF.

General Site History

Manufacturing began in the mid-1880s when Union Tool, a well-drilling equipment manufacturer, began operations on a two-hectare section in the north part of the current REF footprint. The Lindsay Light and Chemical Company (Lindsay) acquired the site in 1931. Lindsay began extracting thorium and rare earths from monazite ore in 1932. From 1932 to 1973, the REF processed approximately 136,000 metric tons of ore. Thorium ore (monazite and bastnaesite sands) was processed at the REF from 1932 to 1963. The plant was shut down in 1973.

Milling and Manufacturing Processes

During the REF operational period (1932 through 1973), various chemical processes were used to produce thorium and rare earths compounds. During the early years of the REF, thorium and rare earths compounds were extracted using a "hard pot" process. Mud residues resulting from this were disposed of at the REF.

In the late 1930s and 1940s, mesothorium (radium-228) was extracted from the black mud residue. Hydrofluoric acid was produced during the war years (the 1940s) by reacting fluorspar, a calcium fluoride mineral, with sulfuric acid. The waste residue from this operation was mostly gypsum.

Between 1954 and 1963, thorium nitrate was produced using the prevailing new acid-cracking technology called the "soft pot" process. The soft pot process involved digestion of the ore using a weak sulfuric acid that resulted in a soft pot cake. The soft pot cake was then leached in refrigerated tanks to produce a thorium solution rather than the solution of rare earth. The rare earths in the resulting residue were crystallized and later purified by additional leaching and chemical treatment. After 1963, the use of monazite ore was limited. The plant started processing bastnaesite, a fluoro-carbonate rare earths ore that contained no thorium. The REF used this process until operations ceased in 1973.

Past Waste Management Practices

Past waste management practices at the REF included on-site storage of unprocessed residues and mud, tailings, scrap metal, and other wastes. Various wastes, including unprocessed residues and muds, tailings, and liquid effluents, were disposed of in settling ponds and waste piles at the REF. Solid waste components included non-ore waste rock and unreacted ores, barium sulfate, and insoluble rare earths and thorium compounds.

Potential Sources of Uranium

The acidic and caustic wastes and slurries, discharged during the manufacturing operations, interacted with the surrounding soil and groundwater to form a variety of minerals and mineral complexes. Elements such as uranium and other metal impurities were likely entrained within the above mineral precipitates as either carbonate, phosphate, or oxide-hydroxide mineral variants or as substituted metals (*e.g.*, uranium replacing magnesium in a dolomite crystal). It is highly plausible that the precipitated minerals and mineral compounds were deposited in the soil matrix and adsorbed onto the soil particles. Similarly, uranium and other metals were also deposited in the soil matrix and adsorbed onto soil particles as an ion or mineral complex.

Previous Remediation and Treatment Activities

Remediation and investigation activities were undertaken from 1997 to 2015 to address the resulting environmental impacts. Sediments and soil were excavated and, depending on physical characteristics, disposed of either at an off-site landfill or on-site after treatment. The remediated soil used as on-site backfill met the soil cleanup criteria but continues to leach uranium at levels above the GWPS.

The majority of contaminated soils were excavated from shallow sand and gravel stratigraphic unit, a glacial outwash deposit primarily composed of limestone and dolomite gravel, sand, and silt. The thickness of the surficial fill material varied between 1.5 and 4.6 meters (m) and contained high amounts of clays, silts, and organic material in addition to imported commercial aggregate such as crushed limestone and granite ballast. The surficial fill may have contained scattered areas of spilled waste material and ore.

Impacted surficial fill material containing a significant fraction of sand and gravel was treated with soil washing technology. During soil washing, the soil was disaggregated in log washers and then wet-screened to remove fine sand, silts, and clays to reduce radiological contamination in the soils. The materials retained on the filter screens, primarily medium and coarse sands and gravels, were tested to comply with uranium and radium's applicable soil cleanup standards. Materials that met the soil cleanup standards were used to backfill previously excavated areas at the REF.

The Pilot-Scale Study

Pilot-Scale testing began in November 2018 and ended in March 2020. It consisted of three phases: the leaching phase, the geochemical stabilization phase, and the confirmation testing phase. The first phase ran from November 2018 to June 2019. The second and third phases lasted from June 2019 through July 2019 and from July 2019 through March 2020.

Specific objectives of the Pilot-Scale Study were as follows:

- a) Assess leachate application, circulation, and extraction issues that may impact full-scale implementation.
- b) Test a higher strength leaching solution compared to previous bench-scale tests.
- c) Confirm the previous bench-scale tests and assess the required number of PVFs for full-scale operations.

The Pilot-Scale Study tested the ISL technology on an approximately 1,500 times larger scale than prior bench-scale studies undertaken at the site.

LITERATURE REVIEW AND FINDINGS

According to the *Manual of Acid, In-Situ Leach Uranium Mining Technology* prepared and published by the International Atomic Energy Agency, ISL technology is innovative and relatively young. The ISL uranium mining technology was developed independently in Russia and the United States in the early 1960s. Although the acid leach system became more popular in Russia, the alkali leach system, primarily a carbonate-based system, became popular in the United States. [1]

In a study, Kim et al. successfully used moderate concentrations of sulfuric acid to leach 90% to 94% uranium from Korean black shale ore. The study also investigated the effects of leaching duration and temperature, particle size and density of the ore, and acid concentration used in the leaching process. [2]

In another study, Ryon et al. found nitric acid to be effective in leaching radium and thorium from uranium ores and tailings. The primary focus of the study was on the effectiveness of the removal of radium and other radionuclides in milling wastes through the application of nitric acid. Tests showed that removing up to 98% of radium and a similar fraction of thorium from uranium ores and tailings was achievable in a two-stage leaching process using a hot 3.0M nitric acid solution. [3]

Kulpa et al. demonstrated the applicability of sodium carbonate- and sodium bicarbonate-based leaching solutions for extracting uranium from contaminated soils. At the RMI Decommissioning Project in Ashtabula, Ohio, operations of a two-ton per batch soil washing plant demonstrated the viability and effectiveness of a 0.2M sodium carbonate and sodium bicarbonate solution. [4]

Zhou and Gu also examined the efficacy of uranium extraction from contaminated soil using sodium carbonate and bicarbonate solutions. The study showed that an increase in bicarbonate concentration increased uranium leaching and that even a small addition of bicarbonate solution to contaminated soil could result in significant uranium leaching. The leaching process was more effective under aerobic conditions than anaerobic conditions, and the leaching process was more effective with a leaching solution with a higher pH value. [5]

At the Fernald Site in Ohio, Buck et al. characterized uranium phases in contaminated soils before examining the carbonate heap leaching technique. The carbonate heap leaching process successfully leached most of the mobile uranium phases from the Fernald soils. Moreover, the addition of bicarbonate to the leaching solution aided in uranium removal by preventing re-precipitation of the dissolved uranium. [6]

In a study, Mason et al. examined the heap leaching process using a carbonate and bicarbonate solution to remove uranium from soils at the Fernald Site. The results showed no significant difference in uranium removal between potassium carbonate and potassium bicarbonate and sodium carbonate and sodium bicarbonate. Additionally, the application of sodium peroxide, an oxidizing reagent, had a significant effect on uranium removal. [7]

Among the acids, sulfuric acid has demonstrated a high leaching performance at a lower cost. Nitric acid, with its high oxidation potential, is considered the most capable leaching agent for uranium. However, its high cost and propensity to impact groundwater with nitrates offsets its leaching advantages relative to sulfuric acid. [8]

Generally, oxidizers are not effective in acid environments. Acid consumption is generally determined by the interaction of an acid with non-uranium minerals of the host media. The acid method becomes uneconomical as the carbonate content increases because of increased acid consumption (that is, each percent of calcium carbonate will consume 1% of sulfuric acid during a complete reaction).

The literature review indicates that alkali-based leaching solutions do not chemically react with limestone aggregate as aggressively as acid-based leaching solutions. They require a longer residence time, and hence more flushes, for leaching compared to acid-based solutions.

The carbonate leaching process is a well-established uranium leaching extraction method. It is based on forming very soluble and stable uranium tricarbonate complex in the presence of excess carbonate and bicarbonate ions in water. The uranium tricarbonate compound has a low adsorption affinity to soil and organic material and can only be formed with hexavalent uranium (*i.e.*, U+6). As such, the carbonate leaching process will typically require the oxidation of tetravalent uranium (*i.e.*, U+4) first in reducing conditions.

METHODS AND MATERIALS

The Pilot-Scale Study consisted of three phases including a leaching phase, a neutralization phase, and a confirmation testing phase. The leaching and neutralization phases of the pilot-scale test were conducted in an in-situ test cell located within a heated 12.2 m x 14.6 m fabric building to maintain a controlled testing environment. The test cell was approximately 11.8 square feet (ft^2) in area and 6.25 m deep, encompassing about 60 cubic meters (m^3) of impacted soil. The sheet piles penetrated the underlying clay layer, located 5.2 m to 5.5 m below the ground surface. The purpose of the sheet piles was to minimize groundwater infiltration and leachate exfiltration from the test cell. The test cell included four six-inch extraction wells, each equipped with a two-hp submersible pump. The extraction wells were used to circulate the groundwater and leachate in the test cell continuously. The test cell was also equipped with one internal and three external two-inch piezometers for monitoring leachate losses and contaminant migration during the testing period. All wells and piezometers were installed to the top of the clay layer. FIGURE 2 illustrates the layout of the test cell, including the key components. FIGURE 3 is a photograph of the Pilot-scale building and the test cell. FIGURE 4 shows the test cell arrangement and the typical piping layout for leachate circulation.



Fig. 2 – Pilot-Scale Study Test Cell Layout



Fig. 3 – Pilot-Scale Study Building and Test Cell



Fig. 4 – Typical Piping Layout of the Test Cell

Two high-level and two low-level liquid level pump controllers were installed in the test cell to automate the operations of the submersible pumps when the leachate levels dropped below a certain level in the event of a pipe leak or if leachate rose too high, respectively. The two high-level float switches were set approximately 37 centimeters (cm) above the soil surface, and two low-level shut-off float switches were set at about 15 cm above the soil surface.

The design Pilot-Scale leaching solution was a mixture of approximately 0.6 molar (M) sodium bicarbonate and 0.23M sodium carbonate. The composition of the solution was based on the results of previous treatability studies. It entailed 1.5 times higher sodium bicarbonate and sodium carbonate molar concentration in the same chemical ratio tested in prior bench-scale studies. The leaching solution was prepared by mixing dry sodium bicarbonate and sodium carbonate with site groundwater in the test cell. Before introducing the chemicals, the test cell was filled with approximately 11,350 liters (L) of groundwater, which was circulated in the test cell for one hour using a two-hp submersible pump in one of the four extraction wells. The groundwater was pumped from on-site monitoring well with a three-inch transfer pump through one of four test cell discharge pipes equipped with an in-line flow meter/totalizer. The leaching solution took three to four days to reach its maximum strength and full leaching potential.

During the leaching phase, the entire 5.2 m of soil column was saturated and treated. The leaching solution was continuously circulated within the test cell to ensure uniform distribution throughout the soil matrix and prevent channeling and vertical stratification. Continuous circulation also facilitated the leaching solution's inward and upward gradient that likely minimized leachate exfiltration and groundwater infiltration. Additionally, one foot of leaching solution was maintained on top of the soil being treated. The leaching solution on top and within the pore space of the soil column constituted one pore volume. The operating fluid depth in the test cell was approximately 5.5 m and was about 1.2 to 1.5 m higher than the surrounding groundwater table.

The uranium leaching process was a batch process that entailed filling the test cell with one pore volume of leaching solution at a given time. The leaching solution was then allowed to remain in contact with the soil and continuously circulated for 30 to 49 days. Depending on the results of leachate analysis, the pregnant leaching solution or leachate was flushed and replaced with a fresh batch of leaching solutions. The batch process was repeated five times, with each flushing and replacement cycle constituting one PVF.

The actual operating concentration of the leaching solution varied from the design concentration due to rounding errors, incomplete purging of leachate volume from previously completed PVF, and leachate losses from the test cell. The highest leachate losses occurred during holidays when circulation of the leachate was paused. Leachate losses, ranging from approximately 43 liters per day (LPD) to 146 LPD, were replenished weekly with site groundwater to maintain a foot of leachate above the soil surface. The volume of water added ranged from approximately 1420 L per PVF to 4088 L per PVF. The addition of water gradually diluted the leaching solutions through the PVF operating period, resulting in marginal dilution of sodium bicarbonate and sodium carbonate concentrations.

During the initial commissioning of the test cell, all four submersible pumps in the extraction wells were operated to yield a cumulative flow rate of approximately 1514 liters per minute (LPM). However, the high extraction rate resulted in the rapid rise of the leaching solution in the test cell due to a slower than anticipated infiltration rate. As such, only one pump was operated to balance the extraction and infiltration rates. An optimal circulation rate of 530 LPM was used to maintain a stable leachate level in the test cell during the first PVF. Each successive PVF yielded less infiltration, most likely due to the accumulation of undissolved chemicals, precipitates, and fine aggregates on the soil surface.

Upon completion of five PVF's, the treated soil in the test cell was neutralized with acidified groundwater. Additionally, two small batches of the treated soil, excavated from the test cell after completion of PVF-03 and PVF-05, were separately neutralized in six 208 L drums. The primary purpose for neutralizing the treated soil in separate 208 L drums was to facilitate confirmation testing, where a controlled environment devoid of test cell leaks and groundwater influx was necessary. The six 208 L steel drums, D-01 through D-06, were retrofitted with two-inch well screens and equipped with submersible pumps to circulate water within the drums. FIGURE 5 depicts the setup of the drums used for neutralizing the treated soil.



Fig. 5 – Confirmation Test Equipment

The treated soil in the drums was neutralized with groundwater acidified with 37% volume-to-volume (v/v) HCL solution. The treated soils in drums D-01 and D-02, collected after PVF-03, were neutralized with 2,050 milliliters (mls) and 1,450 mls of acid. Neutralization and flushing of treated soil in drums D-01 and D-02 soil lasted for 73 days, over three cycles. The soil in drums D-03 through D-06, collected after PVF-05, was flushed and neutralized with approximately 300 L of groundwater and 2,190 mls to 2,400 mls of HCL. The process was completed in 46 days and three flush cycles. After neutralization was complete, soil in all the drums was flushed with fresh groundwater to remove any minor residual sodium bicarbonate and sodium carbonate.

Confirmation tests were conducted to test the leachability of residual uranium from treated and neutralized soil contained in the six 208 L drums. The purpose of confirmation testing was to determine if uranium concentrations remained below the GWPS of 30 picocuries per liter (pCi/L), or approximately 45 micrograms per liter (μ g/L), when the soil remained in continuous contact with site groundwater. Confirmation tests entailed saturating the treated and neutralized soil in the six 208 L drums with site groundwater. The groundwater in the drums was periodically circulated using submersible pumps. Only the volume drawn for sampling purposes and lost through evaporation was replenished to allow residual uranium to accumulate in the water over a long period and thus simulate the condition of slow-moving groundwater through the REF. Confirmation testing of soil in drums D-01 and D-02 was discontinued after two weeks because uranium GWPS were not met. The soil in drums D-03 through D-06 was tested for approximately eight months.

A detailed sampling and analysis program was implemented to measure and monitor the outcome of each phase of the Pilot-Scale Study. During the leaching phase, samples of groundwater circulated in the test cell were collected at the beginning of each PVF to determine baseline groundwater constituent concentrations. Thereafter, weekly leachate samples were collected after the introduction of the leaching solution in the test cell. Finally, samples were collected at the end of each PVF before purging the impregnated leaching solution into the temporary storage tank.

In order to monitor the migration of leachate from the test cell, weekly groundwater samples were collected from piezometers surrounding the test cell. Additionally, groundwater pH was monitored with data loggers installed in each of the three external piezometers. The data loggers were programmed to collect data every two minutes.

Post-neutralization leachate samples were collected from drums D-01 and D-02 before purging and flushing the neutralized soil with site groundwater. During the confirmation phase, groundwater in drums D-03 through D-06 was sampled every 40 days for approximately eight months to measure uranium accumulation in the drums. In all phases, samples were collected in laboratory-provided sample containers and analyzed for parameters listed in TABLE I. Table I also shows the sampling and frequency of each parameter.

	Frequency				
Analytical Parameter	Beginning of PVF	7-Days after leaching Solution Fill	Weekly Sample	End of PVF	
Radiological					
Gross Alpha, Gross Beta, Ra-226, Ra- 228, Th-228, Th-230, Th-232, Pb-210, U-234, U-235, U-238	X	X		X	
Total Uranium	Х	Х	Х	Х	
Metals					
Arsenic, Barium, Boron, Cadmium, Calcium, Chromium, Cobalt, Copper, Iron, Lead, Magnesium, Manganese. Mercury (Low Level), Molybdenum, Nickel, Nickel, Potassium, Selenium, Silver, Sodium, and Zinc	X	Х		Х	
Mercury (Low Level)	Х	Х	Х	Х	
Other Non-radiological					
Chloride, Fluoride, Nitrate, Total Organic Carbon, pH, Ammonia, Nitrites, Orthophosphate, Phosphorus, Total Dissolved Solids, Dissolved Oxygen	X	X		X	
Sulfate and Alkalinity	Х	Х	Х	Х	

 TABLE I – Summary of Groundwater and Leachate Sampling and Analysis Program

Samples of treated and neutralized soil were collected at four different locations within the test cell. At each location, aliquot samples were collected at different depth intervals and composited. The samples were then crushed and uniformly mixed to ensure representativeness. Uranium results ranged between 5.2 micrograms per gram (ug/g) and 8.1 ug/g, with an average of 6.49 ug/g. An average initial uranium concentration of 14.22 ug/g was derived by adding back the uranium removed via leaching.

DISCUSSION OF RESULTS

Leaching of Uranium

The leaching solution successfully leached a significant mass of uranium from the impacted soil. The results were consistent with previous treatability studies completed under controlled laboratory conditions and confirmed that ISL is a viable treatment option for the REF soil.

The total uranium mass removed from the soil in five PVFs averaged approximately 54% of the initial mass in the soil. Roughly 16% -19% of the initial uranium mass was leached in the first PVF with the reported maximum uranium concentration of 23,000 μ g/L. The uranium mass removed in the four successive PVFs was still significant but declined steadily by roughly 30% after each PVF, indicating a progressively more insoluble uranium mass remaining in the soil. TABLE II summarizes the analytical data for samples collected at the end of each PVF and the mass of uranium removed in each PVF.

Pore Volume Flush (PVF) No.	1	2	3	4	5
PVF duration (days)	30	49	40	31	49
Total uranium concentration in leachate, (ug/L)		16,300	9,640	7,800	6,400
Average weekly concentration, (ug/L)		13,544	8,119	5,246	5,126
Average weekly water loss, (gal)	190	189	263	141	130
Average weekly mass removal through water loss, (g)	121	97	81	28	25
Mass of total uranium removed in each PVF, (g)		1,541	972	781	642
Mass of total uranium removed through water loss, (g)	560	699	467	126	185
Cumulative total uranium mass removed, (g)	2,801	5,042	6,481	7,388	8,215
Total uranium removed from soil during each PVF, (ug/g)		2.11	1.35	0.85	0.78
% Total uranium removed from soil during each PVF, (%)		14.8%	9.5%	6.0%	5.5%
Cumulative % total uranium removed from soil during each PVF, (%)	18.5%	33.4%	42.9%	48.9%	54.3%

TABLE II – Uranium	Concentrations in	Leachate and	Uranium Mas	s Removed
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As illustrated in FIGURE 6, most uranium leaching occurred in the first week of each PVF. In the first week, the leaching was markedly higher than leaching with some relatively dilute sodium bicarbonate-only solutions tested in previous bench-scale studies. After the first week, uranium concentrations in the leachate were approximately 60%-80% of the final concentration reported at the end of each PVF. Uranium concentrations in all PVFs, except PVF-02, increased throughout the PVF, with the final sample having the

highest concentration. The composition and strength of the leaching solution and its contact time with the impacted soil were determined to be the critical factors for maximizing uranium removal. After 21 days of leaching, uranium concentrations, on average, increased by approximately 20%. In three of the five PVFs, the highest uranium concentrations were observed on the final day of the PVF.



Fig. 6 – Uranium Concentrations Trends in the Pilot-Scale Study Leachate

Leaching of Other Constituents

The leaching process resulted in the dissolution of non-uranium radionuclides present in the soil. These radionuclides include gross beta, thorium-228, thorium-230, thorium-232, and lead-210. Gross beta includes beta-emitting radionuclides resulting from the decay of uranium-238 and thorium-232. All reported non-uranium radiological constituents, except for thorium-230, exceeded regulatory effluent discharge limits in one or more PVFs. In general, the concentrations of these constituents declined with each successive PVF, similar to uranium concentrations.

Certain non-radiological constituents were also leached during the leaching process. These constituents, including ammonia, arsenic, cadmium, copper, iron, lead, mercury, nickel, zinc, pH, fluoride, chlorides, sulfates, and total dissolved solids (TDS), exceeded their effluent limits. Like uranium, the concentrations of non-radiological constituents declined in each successive PVF.

Leachate Losses

Leachate losses due to evaporation and subsurface leaks were reported throughout the Pilot-Scale Study period. Losses were more pronounced during extended shutdowns, likely due to the inward and upward groundwater gradient resulting from continuous leachate circulation in the test cell. Leachate losses ranged from 43 LPD to 146 LPD and were replenished weekly to maintain one foot of leachate above the soil surface. The groundwater volume added to replenish the losses ranged from approximately 1420 L to 4088 L per PVF. The addition of groundwater resulted in only marginal dilution of the leaching solution.

Neutralization of Treated Soil

The treated soils and associated leachate/groundwater in the test cell and the 208 L drums were successfully neutralized with groundwater acidified with 37% v/v HCL solution. Subsequent testing of the neutralized soil indicated stable and natural levels of alkalinity and pH values.

Confirmation Testing and Sampling

Confirmation tests indicate that uranium concentrations in groundwater, due to uranium leaching from treated soil, comply with the uranium GWPS. When soil, from which more than 50% of uranium mass was leached (in 5 PVFs), was allowed to remain in contact with the site groundwater for 238 days, uranium concentrations in groundwater samples consistently remained below the GWPS, ranging from 14.9 μ g/L to 34.6 μ g/L.

CONCLUSIONS

The pilot-scale study results demonstrate the viability and applicability of the ISL remedial alternative in treating soil at the REF. The leaching solution successfully leached a significant mass of uranium from the impacted soil. The results, consistent with previous treatability studies completed under controlled laboratory conditions, confirmed that ISL is a viable treatment option for addressing residual uranium in the REF soil. Preliminary cost estimates for using the ISL process indicate a savings of approximately \$148 million in direct and indirect remediation costs compared to the excavation and landfill disposal remediation option.

Confirmation tests suggest that removal of approximately 50% of the uranium mass from the soil is required to meet the GWPS. The ISL during the Pilot-Scale Study removed 54% of the initial uranium mass in five PVFs. Roughly 16% -19% of the initial uranium mass was leached in the first PVF. The uranium mass removed in the four successive PVFs was still significant but declined steadily after each PVF, indicating a progressively more insoluble uranium mass remaining on the soil. Critical factors for maximizing uranium leaching were the composition and strength of the leaching solution and its contact time with the impacted soil.

The leachate generated during the leaching process will require treatment and disposal. Leachate analyses indicate the presence of several non-uranium radionuclides and inorganic constituents, which leached out during the leaching process. Several of these constituents exceeded their corresponding effluent standards and will require appropriate management and treatment as part of the leachate treatment program.

The leaching solution took three to four days to reach its maximum strength and full leaching potential because of the time required to mix the dry chemical components. Full-scale operations should consider alternative methods for preparing and applying leaching solutions.

Accumulation of undissolved leaching solution constituents (sodium bicarbonate and sodium carbonate) on the soil surface required neutralization to halt the uranium leaching process fully. Groundwater acidified with HCL was successful in achieving the desired neutralization.

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