Novel Americium Treatment Process for Surface Water and Dust Suppression Water

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INTRODUCTION

The Rocky Flats Environmental Technology Site (RFETS), a former nuclear weapons production plant, has been remediated under CERCLA and decommissioned to become a National Wildlife Refuge. The site conducted this cleanup effort under the Rocky Flats Cleanup Agreement (RFCA) that established limits for the discharge of surface and process waters from the site.

At the end of 2004, while a number of process buildings were undergoing decommissioning, routine monitoring of a discharge pond (Pond A-4) containing approximately 28 million gallons of water was discovered to have been contaminated with a trace amount of Americium-241 (Am-241). While the amount of Am-241 in the pond waters was very low (0.5 – 0.7 pCi/l), it was above the established Colorado stream standard of 0.15 pCi/l for release to off site drainage waters.

The rapid successful treatment of these waters to the regulatory limit was important to the site for two reasons. The first was that the pond was approaching its hold-up limit. Without rapid treatment and release of the Pond A-4 water, typical spring run-off would require water management actions to other drainages onsite or a mass shuttling of water for disposal. The second reason was that this type of contaminated water had not been treated to the stringent stream standard at Rocky Flats before. Technical challenges in treatment could translate to impacts on water and secondary waste management, and ultimately, cost impacts.

REMEDIATION STRATEGY

Upon discovery of the elevated levels of Am-241 in the A-4 pond, an immediate review of existing facilities and treatment options was undertaken. This evaluation determined that a number of treatment options might be possible but, due to the very low Colorado stream standard of 0.15 pCi/l, no proven methods of treatment for this water were readily available. A number of conventional treatment methods such as direct filtration, chemical precipitation, adsorption methods and reverse osmosis (RO) were evaluated for their potential applicability but all of the available methods were either unproven or could not consistently meet the discharge criteria.

A review of the available treatment literature for the removal of trace radionuclides to these very low levels determined that removal of Am-241 and Pu to these extremely low levels had never been consistently achieved. Since it is known that certain actinide elements form colloidal polymers at low concentrations and neutral pH conditions (Santschi, et al, 2000), the initial emphasis of the evaluation was to use multiple stage submicron filtration to remove the Am-241. Initial results of these tests were not successful and showed almost no removal even at filtration levels down to less than 0.2 microns. Reverse osmosis (RO) was considered a possibility but the resultant RO brines, which would amount to approximately 2 – 3 million gallons, would have to be disposed of off site. Off-site disposal was viewed as problematic from a schedule point of view and would also be very expensive.

In addition to the technical challenges of treatment for this contaminated water source, the development of an effective remediation strategy for the contaminated A-4 pond waters was also constrained by important site criteria. The first was that the A-4 discharge pond, which was the terminal discharge pond for a series of collection ponds for surface runoff waters, was nearing its design holdup capacity. In preparation for typical spring runoff flows, the A-4 pond is emptied in the late winter. If Pond A-4 were to remain contaminated into the spring runoff period, then extraordinary measures would have to be taken to route flows to other ponds for retention and routine sampling prior to release in accordance with site procedures.

All of the technical challenges and specific site criteria led to the conclusion that a different approach to the treatment of this problem was necessary and a crash treatability program to identify applicable treatment techniques was undertaken. The goal of this program was to develop treatment options that could be implemented very quickly and would result in the generation of no high volume secondary waste that would be costly to dispose.

TREATABILITY STUDIES

It was decided that a series of treatability studies should be undertaken to establish if treatment of the contaminated water was possible to meet all of the site discharge criteria or if the site should start evaluating other water management alternatives and impacts. Since no known methods of water treatment were identified to treat to the very low limit of less than 0.15 pCi/l, a series of bench-scale treatability tests was performed to develop a method to treat these waters to these extremely low discharge levels.
A literature evaluation of treatment processes and experience on americium removal was initially conducted. It was determined that treatment of water containing very low activity of Am-241 (0.5 to 0.7 pCi/l) was not documented in available research or full-scale project literature. The only promising technology that was historically identified for low-level americium removal was the use of bone char contact columns as the treatment process (Elvin, 2004, Smith, et al, 1998). Since no technical adsorption capacity values were known to be available, it was assumed that the use of bone char would require a very large amount of the adsorbent and the availability of an adequate supply of bone char was uncertain. In addition, the lead contractor for pond operations and the americium removal project was not convinced that treatment beyond simple filtration would be necessary to meet the surface water criterion. These uncertainties pointed out the need for a comprehensive treatability study to define a treatment process or processes for implementation at Rocky Flats.

In support of the expedited treatment project, a series of laboratory-scale treatability tests were conducted, with the following objectives:

- Determine whether the particle size of Am-241 in the pond water would be amenable to direct filtration;

- Assess whether bone char filtration treatment could be used to achieve the 0.15 pCi/l criterion for Am-241 and establish the amount of bone char that would be required to treat approximately 30 million gallons;
- Examine chemical coagulation and flocculation as a means to enhance filterability and removal of Am-241; and
- Explore a novel treatment technique to produce calcium phosphate co-precipitation of Am-241 as a means of simulating the bone char removal process.

The calcium phosphate co-precipitation testing objective was included based upon the results of previous studies in which various other forms of apatite (calcium phosphates) were shown to be comparable to bone char for removal of Pu, Am and U from water (Smith, et al, 1998; Blane & Murphy (undated); and Bostick, et al, 1999).

Pond Water Quality

Routine monitoring and follow-up confirmatory monitoring of the pond in November 2004 showed Am-241 values of 0.565 and 0.563 pCi/l. Another pond upstream of the terminal pond had an Am-241 activity of 0.635 pCi/l. These analyses were obtained on unfiltered 1.9 liter samples and were blank-corrected, the usual methodology for obtaining very low activity radionuclide results. The 2-sigma measurement uncertainty was consistently ±25 percent or less, tracer recoveries were 70 percent or higher, and the reported limit of detection was 0.02 to 0.03 pCi/l. This protocol for Am-241 laboratory analysis was maintained for all treatability samples to achieve consistent precision and accuracy.

The range of Am-241 activity, including bulk samples collected for the treatability studies, and general quality parameters in the pond water are given in Table I.

Table I. Pond A-4 Water Quality

Americium-241	Total Suspended Solids (TSS)	Total Dissolved Solids (TDS)	Total Organic Carbon (TOC)
0.563 – 0.665 pCi/l	6 – 8 mg/l	750 – 1100 mg/l	8 – 9 mg/l

Filtration Tests

A series of filtration tests were conducted on the raw (unconditioned) pond water. Apparatus for the study included a small pump for filter pressurization and a range of filter pore sizes, provided by deep cartridge filter elements for the larger pore sizes (1 to 20 microns) and sealed membrane filters for the smaller sizes (0.2 and 0.45 micron). Each filter run lasted long enough to generate about 4 liters of filtrate, allowing for laboratory analysis of both the filtrate and the filtrate spiked with a tracer. Table II shows the Am-241 results for an unfiltered pond water sample collected from the same bulk sample container, together with the results for each filter pore size.

Table II. Filtration Study

Test Description		Am-241 (pCi/l)
Unfiltered Pond Water		0.665
Filtered Samples:	20 micron	0.669
	10 micron	0.618
	5 micron	0.525
	1 micron	0.630

0.45 micron	0.554
0.2 micron	0.546

These results showed limited removal of Am-241 by filtration for pore sizes less than 20 microns. Considering that removals were only 4 to 21 percent of the unfiltered pond water Am-241 value and that the 2-sigma uncertainty values for the analyses range up to 25 percent, the Am-241 was apparently in solution or in an extremely small colloidal form that was shown to be non-filterable. The fact that significant Am-241 removal was not achieved indicates that direct filtration would not be an effective treatment method, even using commercially available micro-filtration membrane modules, which typically have a pore size of about 0.1 – 0.2 micron. If Am-241 is present as colloidal particles, then the size gradation of those particles lies entirely or mostly below 0.2 microns. It was concluded that direct filtration could not achieve the desired 0.15 pCi/l discharge criterion.

Bone Char Tests

Two grades of bone char supplied by Brimac Carbon Services, Ltd, Greenock, Scotland, UK, were tested in column studies: coarse 8/24 and fine 20/60 mesh sizes. Commercially available bone char consists primarily of heated, milled animal bones and consists of approximately 75% hydroxyapatite, 10-15% amorphous carbon, and 7-9% calcium carbonate. The same bulk pond water sample used in the filtration study was used for the bone char tests. These tests were performed in one-inch diameter columns with peristaltic metering pumps and had a media height of one foot and a bed volume (BV) of 154.4 cubic cm. The initial flowrate through the columns ranged between 40-60 ml/min (Elvin, 2005). Although the pumps were originally set to feed pond water to both the fine and coarse bone char filters with a residence time in the media of about 3 minutes, pores in the media quickly became clogged with fines and algae, which increased the residence times. Over the course of the 200 bed volume test, the mean bed residence time was 4.2 minutes for the coarse bone char and 8.8 minutes for the fine bone char. Effluent samples were taken at regular intervals to determine initial treatment efficiency and look for breakthrough, defined as the treated volume capacity of the media at the point where the effluent activity level equals the discharge criterion of 0.15 pCi/l. The results for three test intervals are shown in Table III.

Table III. Bone Char Column Study

Test Description		**Am-241 (pCi/l)**
Coarse Bone Char	50 Bed Volumes (BV)	0.169
	100 BV	0.330
	200 BV	0.250
Fine Bone Char	50 Bed Volumes (BV)	0.018
	100 BV	0.017
	200 BV	0.041

These results show that the coarse bone char could not reliably meet the required discharge criterion with any of the column run times analyzed. The results for the fine bone char data indicated that the discharge criterion can be met. Breakthrough to the level of the discharge criterion did not occur within 200 bed volumes treated by the fine bone char, although the apparent increase in effluent Am-241 from 100 to 200 bed volumes suggests the beginning of a breakthrough curve. Although not confirmed, it is estimated that column capacities in excess of 500 BV could be achieved before breakthrough using the fine bone char.

Coagulation/Flocculation Tests

Assuming that Am-241 was present in the pond water as a dissolved species or as a very fine, dispersed colloid, a series of coagulation and flocculation tests were conducted to determine if chemical conditioning of these colloidal solids would allow for removal by settling and filtration. Both organic polymers and primary coagulant chemicals were used for coagulation and organic polymers were used for flocculation of the pond water, as shown in Table IV. The first two tests employed low and high molecular weight polymeric coagulants, with a low charge anionic polymer flocculant. The third and fourth tests utilized the addition of ferric chloride and lime, together with the anionic polymer flocculant to produce chemical co-precipitation with ferric hydroxide. The final test entailed the use of lime and a cationic polymer flocculant. All tests were conducted using rapid mixing for chemical addition, slow mixing and settling periods, and 0.45 micron final filtration. Test descriptions and results are shown in Table IV.

Best floc formation and settling were exhibited with ferric chloride addition at 10 ppm. Of several polymer coagulants tested, the high molecular weight Zetag 7571 showed best particle formation. However, none of the coagulation/ flocculation tests resulted in significant Am-241 removal.

Table IV. Solids Coagulation/Flocculation Study

Test Description[A]		**Am-241 (pCi/l)**
Coagulant	**Flocculant**	
Low MW – Zetag 7125, 10 ppm	Low charge anionic – MF[B] E32, 2 ppm	0.530
High MW – Zetag 7571, 10 ppm	MF E32, 2 ppm	0.600
Ferric Chloride, 10 ppm; Lime – pH 9.5	MF E32, 1 ppm	0.570
Ferric Chloride – 5 ppm; Lime – pH 9.5	MF E32, 2 ppm	0.590
Lime – pH 9.5	Low charge cationic – Zetag 7869, 1 ppm	0.565

[A] All decanted samples filtered, 0.45 micron, for analysis.
[B] MF = Magnafloc

Calcium Phosphate Treatment Tests

Removal of Am-241 by bone char filtration is thought to involve co-precipitation of Am-241 with calcium phosphate on the surfaces of bone char particles. Tests were conducted to achieve americium removal by creation of a chemical environment that duplicated the chemical reactions thought to occur in bone char columns.

The key chemical constituents of bone char relative to precipitation and adsorption of metal ions from water are calcium and phosphorus. For example, aluminum ions precipitate with calcium and phosphate in the bone char in slightly alkaline conditions to form alumino-apatite. Similarly, americium reportedly co-precipitates with calcium phosphate on the surfaces of bone char particles, effecting its removal from solution. Treatability testing of the same type of conditions in a chemical reaction was therefore conducted with additions of varying concentrations of phosphoric acid and calcium chloride with appropriate pH adjustment. Optimum conditions were identified for co-precipitation of soluble americium (and plutonium, when present) down to less than 0.1 pCi per liter.

A technique was developed to produce a form of calcium phosphate, simulating the removal mechanism of bone char was developed. This method involved the production of precipitated calcium phosphate under a specific set of chemical conditions that would remove the Am-241 by chemical co-precipitation. A series of tests were conducted to assess the efficiency of this co-precipitation removal mechanism by calcium and phosphate chemical additions to the pond water followed by flocculation, settling and filtration, in lieu of bone char filtration. The test descriptions and resultant Am-241 activities for these tests are shown in Table V.

Table V. Calcium Phosphate Treatment Study

Test Description	Am-241 (pCi/l)
Hydrochloric Acid (HCl) to pH 2; Calcium Nitrate & Sodium Phosphate – 40 ppm Calcium Phosphate; Sodium Hydroxide to pH 9; settle/decant	0.006
Same as above; 8 micron Filter	0.058
HCl to pH 2-3;Calcium Chloride & Sodium Phosphate – 40 ppm Calcium Phosphate; Lime to pH 9.5; 8 micron Filter	0.047
HCl to pH 2-3; Calcium Chloride & Sodium Phosphate – 80 ppm Calcium Phosphate; Lime to pH 9.5; 8 micron Filter	0.003
HCl to pH 5.5; Calcium Chloride & Sodium Phosphate – 80 ppm Calcium Phosphate; Lime to pH 9.5; 8 micron Filter	0.517
Phosphoric Acid – 31 ppm; HCl to pH 5.5; Lime to pH 9.5; 8 micron Filter	0.400
Phosphoric Acid – 31 ppm; Ferric Chloride – 22 ppm Fe – to pH 6; Lime to pH 9.5; 8 micron Filter	0.575
HCl to pH 5.5; Ferric Chloride – 44 ppm Fe – to pH 3.5; Lime to pH 9.5; 8 micron Filter	0.193

Four tests were conducted with a soluble calcium salt and sodium phosphate additions after adjustment of pH to 2 – 3, followed by lime addition to pH 9.5, with an additional test at a slightly acidic initial pH of 5.5. Two tests were run with phosphoric acid at the higher pH of 5.5 – 6 and utilized only lime addition to supply calcium. A final test employed ferric chloride addition for co-precipitation at a pH of 3.5 instead of calcium phosphate.

The tests involving an acidic pH of 2 – 3 and calcium phosphate co-precipitation provided removal of Am-241 to less than the Colorado stream standard of 0.15 pCi/l. Chemical additions to produce a calcium phosphate concentration of 40 ppm were just as successful in removal of Am-241 as was 80 ppm. The tests run with a slightly acidic initial treatment pH of 5.5 – 6 were not successful at Am-241 removal, even when calcium phosphate was formed by calcium chloride salt addition at the higher resultant calcium phosphate concentration of 80 ppm. The test of iron co-precipitation with adjustment of pH to 3.5 showed some removal of Am-241, but the discharge criterion was not achieved.

These results indicate that chemical feeds for co-precipitation of calcium phosphate could be used instead of fine bone char filtration for removal of Am-241 from the Rocky Flats pond water. This chemical treatment process would entail three chemical addition steps, beginning with acid pH adjustment, at which point calcium and phosphate could be added to achieve approximately 40 ppm of calcium phosphate for co-precipitation. Then, lime could be added to an alkaline pH endpoint, followed by flocculating, settling and filtration.

SUMMARY AND DISCUSSION OF TREATABILITY RESULTS

Direct filtration tests showed that Am-241 occurs in the pond water either as a very fine colloidal particle or a dissolved species and could not be removed through filtration even with 0.2 micron sized filters.

Bone char filtration is effective in removal of Am-241, but only in the more fine-grained 20/60 media form. Breakthrough of Am-241 to the level of the discharge criterion was not exhibited after 200 bed volumes treated.

Conventional chemical conditioning of the pond water followed by 0.45 micron was unsuccessful in removal of Am-241. These results lend credence to the fact that Am-241 is present in the pond water as a dissolved ion, colloid or as a chemically bound complex.

The calcium phosphate tests showed that the presumed mechanism of Am-241 removal in bone char filtration could be reproduced in solution to efficiently remove Am-241. The sequence of tests show that co-precipitation of Am-241 with calcium phosphate is pH dependent. The only tests that successfully removed Am-241 to below the discharge criterion were those employing an initial treatment pH in the 2 to 3 range. These results suggest that either or both of two chemical changes might have been prominent in allowing for efficient removal of Am-241 from the pond water:

1. Chemical change in the form of Am-241 in acid conditions, e.g., breaking of a soluble complex or change in the ionic form; and
2. Better co-bonding of Am-241 with the aqueous complex, calcium biphosphate ($CaHPO_4$), which forms in the pH 2 – 3 range, than with calcium phosphate ($Ca_3(PO_4)_2$), which forms in near neutral to alkaline pH (Stumm & Morgan, 1981).

The fact that the test using ferric chloride co-precipitation at pH 3.5 achieved partial removal of Am-241 suggests that the first of these two chemical conditions was more important in making Am-241 amenable to chemical treatment. However, additional study would be needed to assess the influence of pH adjustment on removal of Am-241.

The treatability study showed that two different (although chemically-related) treatment processes could be used for americium removal from the Rocky Flats pond water:

1. Fine bone char filtration, and
2. Calcium phosphate co-precipitation.

The latter process requires a three-step process of pH adjustment to pH 2 – 3, calcium phosphate formation, and lime addition for neutralization and flocculation of the precipitates. Additionally, an anionic flocculant aid was shown to optimize solids settling in the co-precipitation treatment system.

PROCESS DESCRIPTION

Based on the results of the treatability studies, an evaluation of the successful treatment alternatives (chemical treatment vs. bone char contactors) was conducted for operability, cost and material availability. Based on the results of these studies a unique treatment method was conceptualized, designed, constructed and operated as a rapid response action at RFETS. Construction, startup and operations crews were rapidly mobilized and successfully treated about 30 million gallons of radioactively contaminated water below the Colorado stream standard of 0.15 pci/l.

The facility was designed for a unique chemical treatment of the principal contaminant, americium 241, as well as other radionuclides (Pu and U) and trace metals. Unit operations included chemical feed systems, three-stage chemical treatment, clarification, sludge storage and dewatering, filtration and effluent neutralization. In order to meet the desired treatment schedule the treatment facility was designed to hydraulically handle 1400 gpm. A block flow diagram of the treatment system is shown in Figure 1.

The treatment process was designed using the developed chemical reaction system. A process flowsheet is shown in Figure 2. The designed treatment process consisted of seven (7) steps:

1. pH reduction and calcium phosphate formation;
2. Co-precipitation of americium with calcium phosphate, and lime addition;
3. Coagulant addition and flocculation;
4. Solids removal by settling;
5. Bag and cartidge filtration;
6. Polishing with bone char; and
7. pH adjustment prior to discharge.

In the first stage, water was pumped from Pond A-4 at a maximum of 1,400 gpm to a static mixer where the near-neutral influent pH was reduced to 2.5 by addition of sulfuric acid. It is believed that the pH reduction breaks carbonate or other americium complexes and reacts to promote a co-precipitation reaction. Next, phosphoric acid was added followed by calcium chloride in order to produce a calcium phosphate mixture.

In the second stage, a lime slurry was added to increase pH to 9.5, producing a calcium phosphate precipitate.

In the third stage, a polymer coagulant was added to break any colloidal suspensions and promote floc formation.

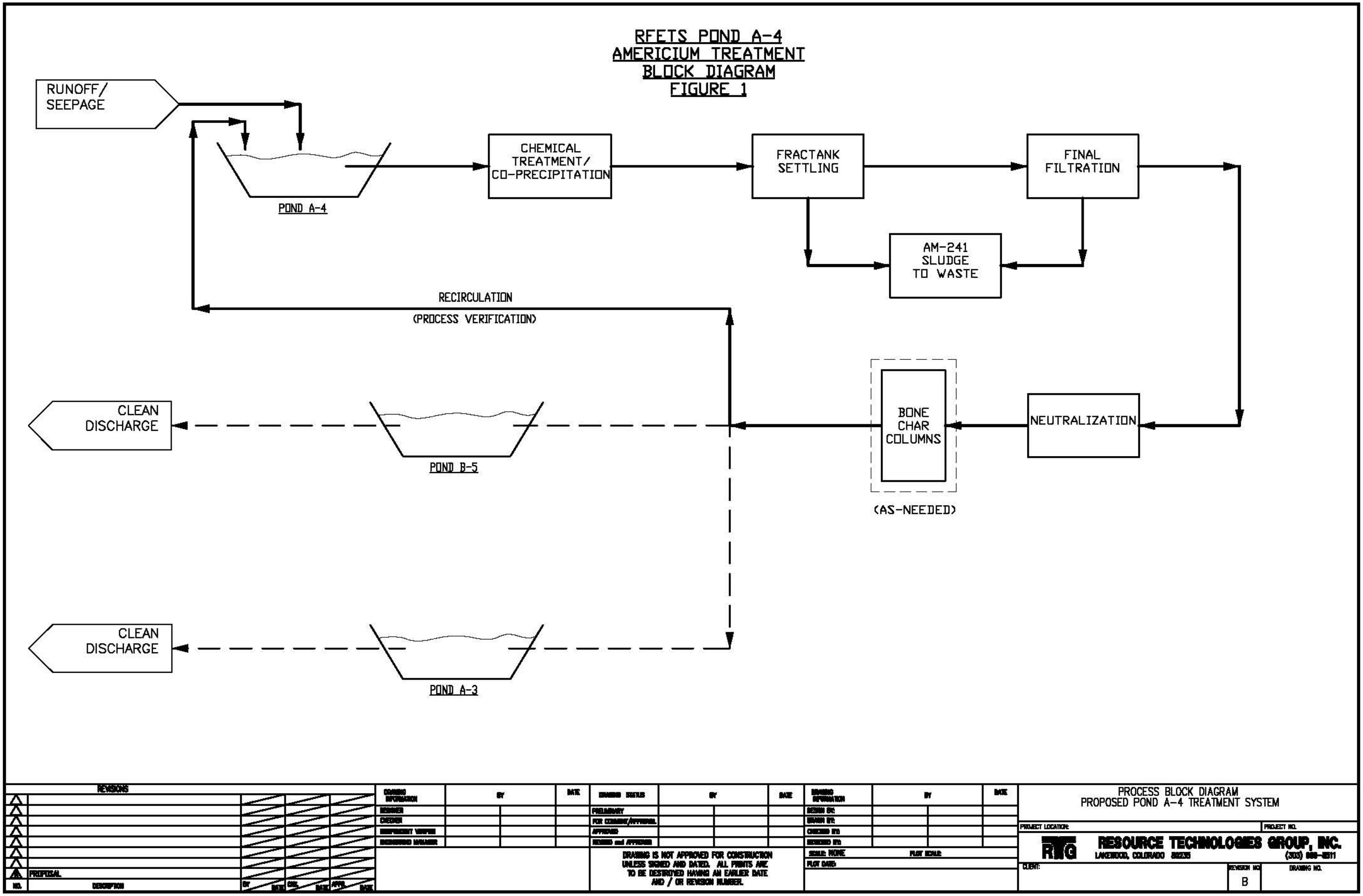
The fourth stage of the process was for removal of the flocculated solids by settling. The process stream was split into two parallel trains. Each train consisted of three inline 25,000 gallon Baker settling tanks where the water flowed by gravity. The first tank provided time for the floc to form by slow and gentle contact. The second tank provided the residence time necessary for settling and concentration of the floc. The third tank provided surge capacity for pumping of the clarified water to the filtration process. The settled floc containing calcium phosphate and americium was collected in the settling tank and transferred to a sludge holding tank. Sludge from the sludge holding tank was then transferred to a plate and frame filter press for dewatering, which produced a filter cake for disposal.

Early experience indicated that while the chemical reaction section of the plant was capable of producing 1,400 gpm, the latter part of the plant (settling and filtration) could not keep up with the design processing rate and the plant flowrate was reduced to a range of 600-800 gpm with excellent results.

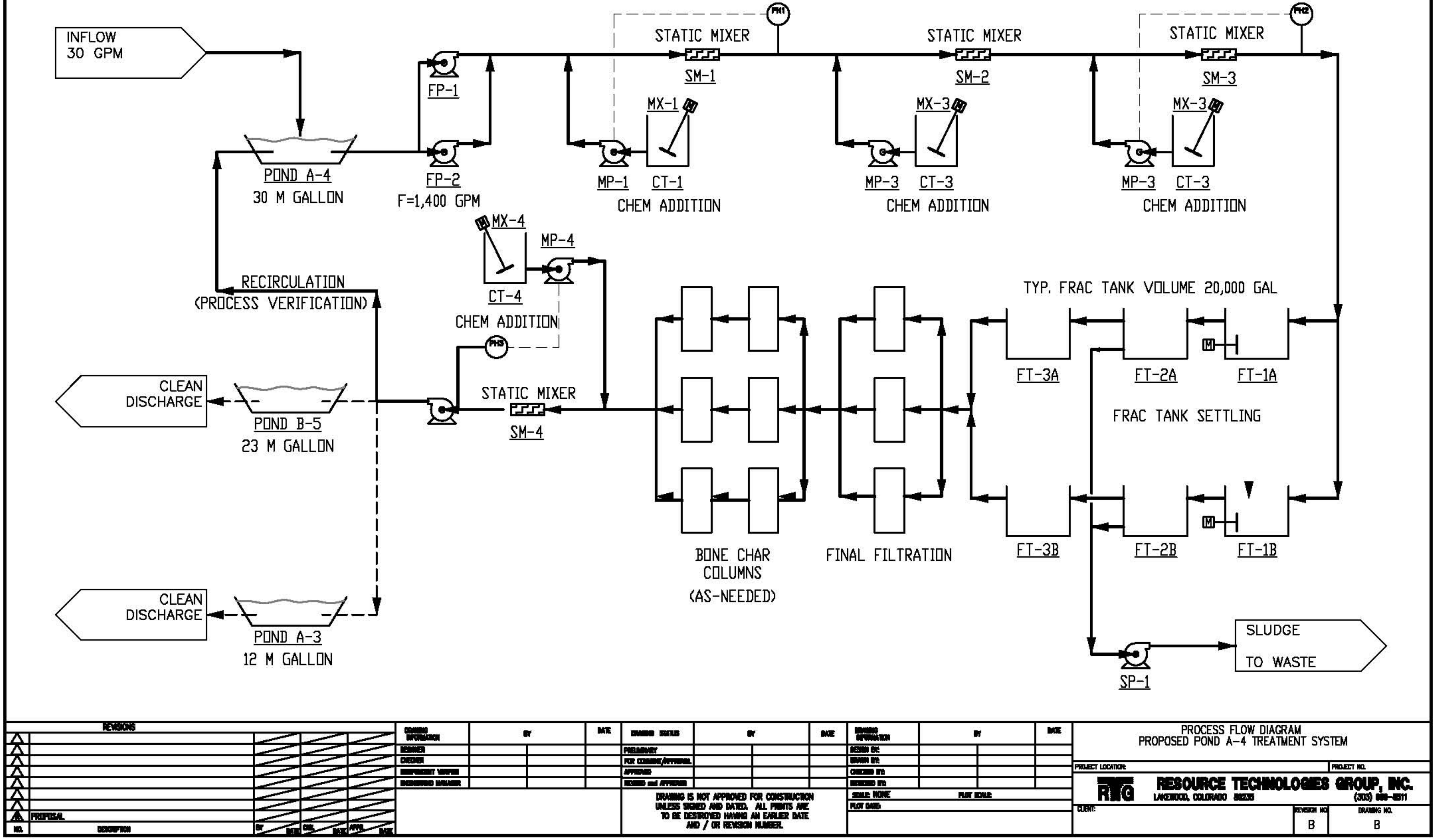
In the fifth stage the supernatant from the precipitation process was passed through a series of filter media. These filters removed any potential carry over of solids from the settling process. The filters consisted of sand filters followed by micron bag and cartridge filters. Backwash from the sand filter was sent to the settling tanks.

At the sixth stage the supernatant was polished in a column loaded with a mixture of fine and coarse bone char. The bone char columns were added as a polishing step since it was demonstrated in the treatability studies that bone char could effectively remove americium.

The final stage of the treatment process re-combined the parallel trains in a common header at which point a final pH adjustment from 9.5 to 8.0 was made in an in-line static mixer with the addition of sulfuric acid. After the in-line pH adjustment, the effluent was sent to Pond A-3 or B-5 for discharge or circulated back to Pond A-4 if the effluent analysis was suspect.


RFETS POND A-4
AMERICIUM TREATMENT
BLOCK DIAGRAM
FIGURE 1
RUNOFF/
SEEPAGE
POND A-4
CHEMICAL
TREATMENT/
CO-PRECIPITATION
FRACTANK
SETTLING
FINAL
FILTRATION
AM-241
SLUDGE
TO WASTE
RECIRCULATION
(PROCESS VERIFICATION)
BONE
CHAR
COLUMNS
(AS-NEEDED)
NEUTRALIZATION
CLEAN
DISCHARGE
POND B-5
CLEAN
DISCHARGE
POND A-3
REVISIONS
PROPOSAL
DRAWING IS NOT APPROVED FOR CONSTRUCTION
UNLESS SIGNED AND DATED. ALL PRINTS ARE
TO BE DESTROYED HAVING AN EARLIER DATE
AND / OR REVISION NUMBER.
PROCESS BLOCK DIAGRAM
PROPOSED POND A-4 TREATMENT SYSTEM
RESOURCE TECHNOLOGIES GROUP, INC.
B

RFETS POND A-4
AMERICIUM TREATMENT
PROCESS FLOW DIAGRAM
FIGURE 2



PROCESS IMPLEMENTATION AND OPERATIONS

Since the proposed treatment process had never been tested in a production mode, the scale-up of the proposed flowsheet from a lab-scale test unit to a full sized production facility capable of treating 1,400 gpm was undertaken with a great deal of care and redundancy. A team of engineers was charged with the detailed design of each of the treatment steps with the understanding that all systems and equipment must be constructed and mobilized very rapidly in order to meet the desired treatment schedule.

Engineering design, equipment specification and process reviews were completed in a two-week period to facilitate the project schedule. Procurement was performed by a team of procurement specialists who identified all process equipment (or suitable alternates) for expedited delivery. The project team and RFETS management felt that the risk of spring run-off was sufficiently high that all efforts on the project were expedited to enable the processing of as much water as possible before any significant snow melt or run-off occurred.

Treatment Plant Construction

Following process design and equipment selection for all unit processes, accelerated construction was undertaken on an around-the-clock basis. All equipment was identified for immediate delivery and installation and, if equipment was not available, alternate equipment or facilities were constructed to keep the construction schedule on track. All necessary equipment was procured, delivered and constructed within a three-week time frame and operational testing was conducted on each of the separate process sections in parallel to minimize delays. Since the facility was constructed and operated in the middle of winter, the facility design and construction included insulation and winterization to prevent the freezing of pipelines and process equipment.

On a parallel track, since the developed chemical treatment process had never been implemented on a production scale basis, absorption columns for installation of bone char treatment were also added to the process as a polishing step after the chemical treatment process. The bone char columns were loaded with fine and coarse particle size bone char media from the only source available to the US market at the time. The entire U.S. supply of readily available bone char was purchased for immediate delivery to load a parallel train, 2 column bone char absorption system. The columns were configured as a polishing process after the chemical treatment system in the initial installation and startup phase.

The initial monitoring results from the chemical treatment process train showed excellent americium removals to levels well below 0.15 pCi/l of Am-241 with most results in the ND to 0.1 pCi/l range. The bone char polishing absorbers were found to be redundant as the chemical treatment system performed flawlessly. It was also determined that the very fine particle size range of the bone char media caused large pressure drops across the columns, which limited the overall hydraulic capacity of the system, even though the contactors were receiving high-quality, pre-filtered feed from the chemical treatment system.

Treatment Plant Operations

The treatment plant was operated on a 24-hour per day, 7 day per week basis with a team of skilled and trained water treatment plant operators. A crew of four personnel was used on each shift which included a pond pump and pipeline operator, two treatment plant operators and a maintenance/chemical feed operator.

The majority of the equipment operated flawlessly and produced effluent water quality that was consistently well below the discharge criteria of 0.15 pCi/l. Operational difficulties with the sand filters and the static polishing filters were a continued source of maintenance but these problems were overcome through the efforts of a skilled crew of operators and dedication to operational objectives and safety.

The treatment system removed the radioactive americium contaminant in the form of a settled calcium phosphate sludge which was dewatered in a high pressure filter press. The filter cake from this operation was sufficiently dry to allow direct disposal in roll-off disposal containers. Disposal of the contaminated sludges was performed by the site solid waste disposal operations and was sent to an off-site disposal facility licensed for low-level radioactive waste. Quantities of sludge requiring off-site disposal were much lower than would have been required if bone char absorption had been the americium removal process.

Health and Safety

During the entire campaign of treatment the facility was operated without any significant safety issues. This was especially significant due to the rapid pace of construction and operation for this treatment facility.

Treatment Costs

The overall unit treatment cost, including process development/design, capital equipment, construction and operation of the Pond A-4 treatment facility, was approximately $0.20/gallon. In contrast, the site had experienced radioactive liquid waste disposal costs in the range of $10-$13/gallon for off-site transportation and disposal. The overall benefit of implementing this novel calcium phosphate precipitation process was that the Rocky Flats Closure Project saved in excess of $30 million, when compared with alternate treatment technologies (e.g., reverse osmosis treatment, with off-site brine disposal).

BUILDING 776 AND 371 SURFACE RUN-OFF AND DUST SUPPRESSION WATER TREATMENT

As the Pond A-4 treatment campaign was winding down, additional water treatment needs were identified where the novel calcium phosphate treatment process might be employed. During the decommissioning and demolition (D&D) of Buildings 776 and 371, it was determined that a treatment system would be required that would be capable of treating and discharging the surface run-off and dust suppression waters from demolition of these two buildings without having to shuttle water to the onsite treatment facility at Building 891, which had a much smaller capacity than was needed. Based on the recent success at Pond A-4, it was felt that, since the treatment system employed similar chemical treatment processes to the onsite system, a logical step would be to use it to support D&D of Building 776 and 371. This process became a modular extension of Building 891, which allowed for the accelerated demolition for 891. A series of treatability studies was conducted to determine what treatment would be required to be in compliance with the Colorado stream standards for plutonium (Pu) and americium of 0.15 pCi/l for discharge to the site drainage system. It was assumed that water generated in the demolition of these buildings would be potentially contaminated with Pu, Am, and U.

Initial water quality evaluations of the Building 776 surface run-off and dust suppression water indicated that the water exceeded the discharge levels of 0.15 pCi/l for both Am and Pu. In addition, the water exceeded the action levels of several other trace metal species.

Treatability studies were conducted to evaluate the following treatment technologies.

- Direct filtration
- Chemical Precipitation
- Reverse Osmosis
- Coagulation/Flocculation/Settling
- Pond A-4 Calcium Phosphate Co-Precipitation Process

The results of the treatability study on Building 776 dust suppression water indicated that either a multiple component chemical precipitation system with polishing filtration or a reverse osmosis (RO) system could be effective in treating B776 dust suppression water for discharge in accordance with the action levels/standards required in RFCA. However, the variations seen in untreated water radionuclide concentrations would not lend themselves to using direct filtration as the primary treatment process. That variability also necessitated having multiple chemical additions to enhance settling and compliance for the waters tested.

Since radionuclides were removed efficiently with chemical treatment followed by filtration based on the Pond A-4 experience, reverse osmosis (RO) was not recommended. The RO process would potentially generate a high volume waste stream consisting of the reject brine from the RO modules. A relatively high percentage of the initial volume (15 to 25% brine) would require additional treatment or stabilization for disposal if reverse osmosis was selected. Sludge and filter media from multiple stage chemical treatment and filtration would be less than 5% of the initial dust suppression water volume.

After completion of the treatability studies, a process evaluation was conducted to determine if unit processes and equipment from the Pond A-4 treatment process could be modified to augment the fixed treatment plant (Building 891) and achieve the treatment goals for Buildings 776 and 371. This evaluation was performed for the conversion of the americium treatment facility to treat radioactively-contaminated dust suppression water and runoff. Process design changes were completed, key equipment was specified, and construction of a modified treatment facility for the D&D wastewaters was undertaken in the spring and summer of 2005. The modified system included the A-4 Pond chemical treatment steps plus ferric chloride addition for trace metals co-precipitation, but excluding bone char filtration. The system was started up and successfully operated to treat all of the surface run-off and dust suppression waters at these two buildings. About 2 million gallons of D&D wastewater was treated to full compliance with the stringent Colorado stream standards and discharged at the end of the project.

CONCLUSIONS

A novel chemical treatment system was developed and implemented at the RFETS to treat Am-241 contaminated pond water, surface run-off and D&D dust suppression water during the later stages of the D&D effort at Rocky Flats.

This novel chemical treatment system allowed for highly efficient, high-volume treatment of all contaminated waste waters to the very low stream standard of 0.15 pCi/l with strict compliance to the RFCA discharge criteria for release to off-site surface waters. The rapid development and implementation of the treatment system avoided water management issues that would have had to be addressed if contaminated water had remained in Pond A-4 into the Spring of 2005. Implementation of this treatment system for the Pond A-4 waters and the D&D waters from Buildings 776 and 371 enabled the site to achieve cost-effective treatment that minimized secondary waste generation, avoiding the need for expensive off-site water disposal. Water treatment was conducted for a cost of less than $0.20/gal which included all development costs, capital costs and operational costs. This innovative and rapid-response effort saved the RFETS cleanup program well in excess of $30 million for the potential cost of off-site transportation and treatment of radioactive liquid waste.

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REFERENCES

1. Blane, D.E. and E.L. Murphy, undated. Mound Laboratory Studies on the Removal of Radionuclides from Waste Water Using Bone Char, Division of Military Operations, Mound Laboratory, Ohio.
2. Bostick, W.D., R.J. Stevenson, R.J. Jarabek and J. L. Conca, 1999. Use of Apatite and Bone Char for the Removal of Soluble Radionuclides in Authentic and Simulated DOE Groundwater, Advances in Environmental Research, MS 548-99.
3. Elvin, Jerry, 2004. Personal Communication: Discussion of Bone Char Effectiveness in Removal of Transuranics from Water, Brimac Carbon Services, Ltd., Greenock, Scotland, UK, December.
4. Elvin, Jerry, 2005. Personal Communication: Discussion of Bone Char Absorption Kinetics for Column Testing, Brimac Carbon Services, Ltd., Greenock, Scotland, UK, January.
5. Santschi, P.H., K. Roberts and L. Guo, 2000. Final Report on Phase Speciation of Pu and Am for 'Actinide Migration Studies at the Rocky Flats Environmental Technology Site', Texas A&M University, September 29.
6. Smith, C.L., R.D. Busch, M.D. Siegel, and C. Baldwin, 1998. Removal of Metals and Radionuclides using Apatite and Other Natural Sorbents, Technical Completion Report, New Mexico Waste-management Education and Research Consortium (WERC) in cooperation with U.S. DOE, October.
7. Stumm, W. and J.J. Morgan, 1981. Aquatic Chemistry, 2nd Edition, John Wiley & Sons, Inc.