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ALPHA REMOVAL PROCESS FILTER CLEANING RECOMMENDATIONS (U)

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Summary

Defense Waste Processing Facility (DWPF) personnel asked Savannah River Technology Center (SRTC) personnel to provide recommendations for chemically cleaning the Alpha Removal Process (Building 512-S) filters. The authors reviewed previous SRTC filter cleaning experience with bench-scale radioactive filters and pilot-scale simulant filters from tests with simulated and actual waste. From reviewing the previous filter cleaning data and assuming the heel in the 512-S filtration system is 85 gallons or less, the authors recommend the following cleaning protocol for the Alpha Removal Process filters.

1. Fill the surge tank with 500 gallons total of flush water, and perform a single pass flush. The flush water should be 0.02 M NaOH.
2. Fill the surge tank with 220 gallons of flush water and fill the backpulse tank with 280 gallons of flush water. Circulate the flush water through the filter and back to the surge tank for 60 minutes, backpulsing the filter with approximately 70 gallons of flush water every 15 minutes. The axial velocity should be 6 – 9 ft/s and the TMP should be approximately 30 psi. Transfer the flush water to the precipitate tank.
3. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank.
4. Fill the surge tank with 220 gallons of 0.5 M oxalic acid and fill the backpulse tank with 280 gallons of 0.5 M oxalic acid. Circulate the acid through the filter and back to the surge tank for 60 minutes, backpulsing the filter with approximately 70 gallons of acid every 15 minutes. The axial velocity should be 6 – 9 ft/s and the TMP should be approximately 30 psi. Transfer the acid to the precipitate tank. Measure the filter flux during cleaning. If oxalic acid flux is acceptable, proceed to the next step. If oxalic acid flux is not acceptable, repeat this step. The authors recommend a target oxalic acid flux of 0.5 – 1.0 gpm/ft^2 at 30 psi.
5. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank.
6. Fill the surge tank with 500 gallons total of flush water, and perform a single pass flush.
7. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank.
8. Continue filtering waste slurries.

If ARP personnel elect not to measure filter flux during acid cleaning, they risk not knowing whether the filter is clean until after they have flushed the system (and added an additional 1500 gallons of flush water to the system). Therefore, if they elect not to measure filter flux during cleaning, the authors recommend performing the oxalic acid cleaning step three times. In addition, ARP personnel should measure the heel in the filtration system during equipment testing to verify the assumptions made are correct.

Introduction

The Savannah River Site (SRS) is developing a process to treat radioactive waste that is low in cesium-137, but high in strontium-90, plutonium, uranium, and neptunium. The process is the Alpha Removal Process (ARP) located in Building 512-S.

This process adds monosodium titanate (MST) to high level waste supernate. The MST sorbs soluble strontium, plutonium, uranium, and neptunium. The process then filters the resulting slurry, which contains entrained metal hydroxide sludge and MST, to remove the insoluble solids. Operations next wash the concentrated solids to reduce the sodium and nitrite concentrations, and transport the slurry to the Defense Waste Processing Facility for vitrification. The filtrate transfers to Z-area for disposal in a cement-based waste form.

Filter fouling occurs during the process, reducing throughput. Personnel can recover throughput by backpulsing the filter to remove the trapped solids, but chemical cleaning is ultimately required. The current baseline cleaning process employs a 1.66 M sodium hydroxide flush, a 0.0055 M sodium hydroxide flush, a 0.2 M oxalic acid cleaning to dissolve sludge and MST that accumulates on the filter surface and within the filter pores,[1] a 0.0055 M sodium hydroxide flush, and a 1.66 M sodium hydroxide flush.[2] Engineering personnel asked SRTC to provide recommendations for chemically cleaning the ARP filters.[2]

Approach

The authors reviewed previous filter cleaning experience with bench-scale (a single 3/8 inch ID tube, 2 feet long) and pilot-scale (7 filter tubes, 5/8 inch ID, 10 feet long) filters. In addition, they reviewed the previous technical bases reports for the Late Washing Facility.[3,4] These prior documents incorporate lessons learned from operation of "full scale" (144 filters, 5/8 inch ID, 10 feet long) filters for the In-Tank Precipitation process. [While the In-Tank Precipitation experience involved slurries that also contained tetraphenylborate solids, the general protocols remain largely unchanged from the earlier work. The primary filter foulants in the earlier process were also MST and the metal hydroxide sludge of the Actinide Removal Process.] The recommendations therein also derive from reviews from representatives of various cross-flow filter manufacturers and from industrial operators of such units.

SRTC personnel conducted actual waste filtration tests in 2001 to support the Salt Waste Processing Facility.[5,6] During these tests, they cleaned the filter in the following manner. They flushed the filter system two times with 0.01 M sodium hydroxide. Flushing the system with dilute caustic removes solid particles that are not strongly attached to the filter and reduces the free hydroxide concentration from ~ 2 M to ~ 0.01 M. The 0.01 M sodium hydroxide is sufficiently basic to prevent aluminum hydroxide precipitation. The dilute caustic also ensures that minimal free hydroxide is in the heel that remains following the flush. A high free hydroxide concentration in the heel would neutralize some of the acid cleaning solution added and make it less effective. Following the dilute caustic flush, they cleaned the filter with four batches of 0.5 M oxalic acid. [Prior work demonstrated oxalic acid worked better than other alternatives, including nitric acid.[1]] Personnel measured filter flux during cleaning and noticed significant improvement following the third batch of oxalic acid. Following the oxalic acid cleaning, they flushed the system two times with 0.01 M sodium hydroxide. The dilute caustic ensures that the heel is basic prior to the addition of fresh feed slurry to prevent aluminum precipitation.

University of South Carolina personnel performed pilot-scale simulant filtration tests to support the Salt Waste Processing Facility.[7,8] During these tests, they chemically cleaned the crossflow filters as follows.[9] They flushed the filter system with deionized water for 20 minutes, cleaned the filter with 0.1 M oxalic acid heated to 50 ºC for 60 minutes, flushed the system with deionized water for 20 minutes, cleaned the system with 0.2 M sodium hydroxide heated to 50 ºC for 60 minutes, and flushed the system with deionized water for 20 minutes. The filters were cleaned at the end of each test program and at the beginning of the subsequent test program.

Chemical Cleaning Approach after Accounting for Tank Heels

The tanks, equipment, and piping in 512-S will contain heels that will dilute cleaning solutions and reduce the effectiveness of water flushes. Personnel performed calculations to determine the impact of these heels on the cleaning process. They used the following assumptions to perform the calculations.

- The surge tank pump interlock shuts the pump down at a tank level of 50 gallons.
- DWPF procedures shut the surge tank pump down at 75 gallons.
- The pump discharge heel is 10 gallons.
- The filter shell and filtrate piping contain 245 gallons of liquid.
- The surge tank can hold 500 gallons.
- The backpulse volume is 70 gallons.
- Complete backmixing occurs.

To perform calculations, the authors considered three cases.

1. The pump operates until the interlock shuts it down and all fluid in the filter system drains to the surge tank (60 gallon heel).
2. The pump operates until procedural shutdown and all fluid in the filter system drains to the surge tank (85 gallon heel).
3. The filter shell and filtrate tubing drain minimally (245 gallon heel).

Table 1 shows the calculation of insoluble solids, free hydroxide, and oxalic acid following chemical cleaning and water flushes with a tank heel of 60 gallons. The volume of all additions to the surge tank is 440 gallons. The oxalic acid concentration is 0.5 M and the flush water is 0.02 M NaOH. Three water flushes are required to reduce the insoluble solids loading below 0.05 wt %. Because of dilution with the heel, the oxalic acid concentration is 0.44 M. Following oxalic acid cleaning, three water flushes are required to produce a fluid containing greater than 0.01 M free hydroxide. Increasing the free hydroxide concentration following oxalic acid cleaning to 0.1 M would reduce the number of water flushes required to two. Additional oxalic acid cleaning cycles may be necessary to ensure adequate filter flux.

Table 2 shows the calculation of insoluble solids, free hydroxide, and oxalic acid following chemical cleaning and water flushes with a tank heel of 85 gallons. The volume of all additions to the surge tank is 415 gallons. The oxalic acid concentration is 0.5 M and the flush water is 0.02 M NaOH. Because of dilution from heels, the oxalic acid concentration during cleaning is 0.41 M. Three water flushes are required to reduce the insoluble solids loading below 0.05 wt %. Following oxalic acid cleaning, three water flushes are required to produce a fluid containing greater than 0.01 M free hydroxide. Increasing the free hydroxide concentration following oxalic acid cleaning to 0.1 M would reduce the number of water flushes required to two. Additional oxalic acid cleaning cycles may be necessary to ensure adequate filter flux.

Table 1. Surge Tank Composition with 60 gallon Tank Heel

	Insoluble Solids (wt %)	Free Hydroxide (M)	Oxalic Acid (M)
Feed slurry	5.000	2.000	0.000
Flush water	0.600	0.258	0.000
Flush water	0.072	0.049	0.000
Flush water	0.009	0.023	0.000
0.5 M Oxalic acid	0.000	0.000	0.439
Flush water	0.000	0.000	0.044
Flush water	0.000	0.007	0.000
Flush water	**0.000**	**0.018**	**0.000**
Flush water	0.000	0.020	0.000

Table 2. Surge Tank Composition with 85 gallon Tank Heel

	Insoluble Solids (wt %)	Free Hydroxide (M)	Oxalic Acid (M)
Feed slurry	5.000	2.000	0.000
Flush water	0.850	0.357	0.000
Flush water	0.145	0.077	0.000
Flush water	0.025	0.030	0.000
0.5 M oxalic Acid	0.000	0.000	0.412
Flush water	0.000	0.000	0.062
Flush water	0.000	0.000	0.002
Flush water	**0.000**	**0.016**	**0.000**
Flush water	0.000	0.019	0.000

Table 3 shows the calculation of insoluble solids, free hydroxide, and oxalic acid following chemical cleaning and water flushes with a tank heel of 245 gallons. The volume of all additions to the surge tank is 255 gallons. The oxalic acid concentration is 0.5 M and the flush water is 0.01 M NaOH prior to oxalic acid cleaning and 0.1 M NaOH following oxalic acid cleaning. Six water flushes are required to reduce the insoluble solids loading below 0.07 wt %. Because of the high insoluble solids present after six water flushes and dilution from the tank heel, two cleaning cycles were used in this calculation. The oxalic acid concentration is 0.25 M during the first cleaning cycle and 0.38 M during the second cleaning cycle. Following oxalic acid cleaning, four water flushes are required to produce a fluid containing greater than 0.01 M free hydroxide.

If the tank heel is 245 gallons, cleaning the filters is likely to be very difficult and require large volumes of cleaning solution. ARP personnel should measure the heel in the 512-S filtration system during system testing.

Table 3. Surge Tank Composition with 245 gallon Tank Heel

	Insoluble Solids (wt %)	Free Hydroxide (M)	Oxalic Acid (M)
Feed slurry	5.000	2.000	0.000
Flush water	2.450	0.985	0.000
Flush water	1.201	0.488	0.000
Flush water	0.588	0.244	0.000
Flush water	0.288	0.125	0.000
Flush water	0.141	0.066	0.000
Flush water	0.069	0.038	0.000
0.5 M oxalic acid	0.000	0.000	0.246
0.5 M oxalic acid	0.000	0.000	0.375
Flush water	0.000	0.000	0.158
Flush water	0.000	0.000	0.052
Flush water	0.000	0.000	0.000
Flush water	**0.000**	**0.051**	**0.000**
Flush water	0.000	0.076	0.000

Filter Cleaning

Assuming the heel in the 512-S filtration system is 85 gallons or less, ARP personnel should clean the 512-S filters as follows.

1. Fill the surge tank with 500 gallons total of flush water, and perform a single pass flush. The flush water should be 0.02 M NaOH. This step will reduce the insoluble solids loading in the surge tank and reduce the free hydroxide concentration of the slurry.
2. Fill the surge tank with 220 gallons of flush water and fill the backpulse tank with 280 gallons of flush water (the backpulse tank water can be added through several additions). Circulate the flush water through the filter and back to the surge tank for 60 minutes, backpulsing the filter with approximately 70 gallons of flush water every 15 minutes. Transfer the flush water to the precipitate tank. This step further reduces the insoluble solids loading in the surge tank. The backpulses will remove some of the trapped solids from the filter surface.
3. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank. This step will reduce the insoluble solids loading in the surge tank and reduce the free hydroxide concentration of the slurry.
4. Fill the surge tank with 220 gallons of 0.5 M oxalic acid and fill the backpulse tank with 280 gallons of 0.5 M oxalic acid (the backpulse tank acid can be added through several additions). Circulate the acid through the filter and back to the surge tank for 60 minutes, backpulsing the filter with approximately 70 gallons of acid every 15 minutes. Transfer the acid to the precipitate tank. Measure the filter flux during cleaning. If oxalic acid flux is acceptable, proceed to the next step. If oxalic acid flux is not acceptable, repeat this step. Typically, clean water fluxes are 1 – 2 gpm/ft^2 at 20 psi with simulant filter systems and 0.25 – 0.75 gpm/ft^2 at 20 psi with actual waste filter systems.[10] The oxalic acid dissolves iron, MST, and silicon that are trapped in the filter. The backpulses help dislodge the trapped particles. Heating the oxalic acid solution would improve chemical cleaning effectiveness.[1] ARP personnel should attempt to perform this step at 30 – 50 ºC.
5. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank. The flush water will dilute and remove

dissolved solids and hydronium ions. If backmixing is less than ideal, flushing from the shell side will cause the shell side of the filter to become basic.

6. Fill the surge tank with 500 gallons total of flush water, and perform a single pass flush. The flush water will dilute and remove dissolved solids and hydronium ions.
7. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank. The flush water will dilute and remove dissolved solids and hydronium ions. The final flush will ensure both the feed and filtrate sides are basic.

Recommendation

From reviewing the previous filter cleaning data and assuming the heel in the 512-S filtration system is 85 gallons or less, the authors recommend the following cleaning protocol for the Alpha Removal Process filters.

1. Fill the surge tank with 500 gallons total of flush water, and perform a single pass flush. The flush water should be 0.02 M NaOH.
2. Fill the surge tank with 220 gallons of flush water and fill the backpulse tank with 280 gallons of flush water. Circulate the flush water through the filter and back to the surge tank for 60 minutes, backpulsing the filter with approximately 70 gallons of flush water every 15 minutes. The axial velocity should be 6 – 9 ft/s and the TMP should be approximately 30 psi. Transfer the flush water to the precipitate tank.
3. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank.
4. Fill the surge tank with 220 gallons of 0.5 M oxalic acid and fill the backpulse tank with 280 gallons of 0.5 M oxalic acid. Circulate the acid through the filter and back to the surge tank for 60 minutes, backpulsing the filter with approximately 70 gallons of acid every 15 minutes. The axial velocity should be 6 – 9 ft/s and the TMP should be approximately 30 psi. Transfer the acid to the precipitate tank. Measure the filter flux during cleaning. If oxalic acid flux is acceptable, proceed to the next step. If oxalic acid flux is not acceptable, repeat this step. The authors recommend a target oxalic acid flux of 0.5 – 1.0 gpm/ft^2 at 30 psi.
5. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank.
6. Fill the surge tank with 500 gallons total of flush water, and perform a single pass flush.
7. Fill the surge tank with flush water (500 gallons total including heel) from the shell side. Transfer the flush water to the precipitate tank.
8. Continue filtering waste slurries.

If ARP personnel elect not to measure filter flux during acid cleaning, they risk not knowing whether the filter is clean until after they have flushed the system (and added an additional 1500 gallons of flush water to the system). Therefore, if they elect not to measure filter flux during cleaning, the authors recommend performing the oxalic acid cleaning step three times. In addition, ARP personnel should measure the heel in the filtration system during equipment testing to verify the assumptions made are correct.

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