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FILTRATION ENGINEERING STUDY TO UPGRADE THE ETF

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Document Number: WHC-SD-ETF-ES-002, Rev. 0

Document Title: Filtration Engineering Study to Upgrade the EFT

Release Date: 10/18/95

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SUPPORTING DOCUMENT

1. Total Pages 37

2. Title

Filtration Engineering Study to Upgrade the ETF

3. Number

WHC-SD-ETF-ES-002

4. Rev No.

0

5. Key Words

filter
waste water
Effluent Treatment Facility
ultrafilter
fouling
colloids

6. Author

Name: F.N. McDonald

Signature

Organization/Charge Code

86230/A2008
86300/A2049 *ew*

7. Abstract

Filtration technologies are evaluated which have potential to augment or upgrade the 200 Area Effluent Treatment Facility. The study was written in anticipation of treating future waste waters that have high fouling potentials. The three ultrafilters judged to be capable of treating future waste waters are: hollow fiber, tubular, and centrifugal.

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RELEASE STAMP

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DATE OCT 18 1995

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WMC-SD-ETF-ES-002, Rev 0

FILTRATION ENGINEERING STUDY TO UPGRADE THE ETF

Issued by

Westinghouse Hanford Company

for the

U.S. DEPARTMENT OF ENERGY, RICHLAND OPERATIONS OFFICE
RICHLAND, WASHINGTON

October 1995

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WHC-SD-ETF-ES-002, Rev 0

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ABBREVIATIONS AND ACRONYMS

Conc.	Concentration
CPVC	Post Chlorinated Polyvinyl Chloride
D&D	Decontamination and Decommissioning
ETF	200 Area Effluent Treatment Facility
gpm	Gallons per minute
LERF	Liquid Effluent Retention Facility
NMWC	Nominal Molecular Weight Cutoff
PC	Process Condensate generated by the 242-A Evaporator
ppm	Parts per million
psig	Pounds per square inch gauge
PVC	Polyvinyl Chloride
PVDF	Polyvinylidene di-fluoride
Rad	Radioactive
RD&D	Dangerous Waste Research, Research, Development, and Demonstration Treatment and Storage Activity
SRS	Savannah River Site
TRU	Transuranic
TSS	Total Suspended Solids
UF	Ultrafilter
UVOX	Ultraviolet Light Oxidation

Filtration Engineering Study to Upgrade the ETF

EXECUTIVE SUMMARY

Commercially available filtration technologies are evaluated which have the potential to augment or upgrade the filtration capabilities of the 200 East Area Effluent Treatment Facility (ETF). The ETF will treat aqueous waste streams in the future which may foul the filtration equipment presently installed in the ETF. These future waste streams are anticipated to contain colloidal and microbiological components.

To ensure the ETF will be able to treat these future waste streams successfully, the existing ETF fine filter (i.e., the 2-micron rated microfilter) should be replaced with one of three ultrafilters (hollow fiber, tubular, or centrifugal). Such filters should not foul in the presence of colloids and biorganisms.

Alternatively, a decision could be made to add ultrafilter capacity to the existing ETF filters. Problematic aqueous wastes could be treated by valving in the standby filters. If the amount of wastes needing treatment by ultrafiltration were limited (and this has yet to be determined), then the capacity of the ultrafilter could be less than the rated capacity of the ETF.

To protect the ultrafilter, rough filter cartridges rated at between 100 and 400 microns should function as prefilters. The new rough filter cartridges could be installed into the existing rough filter housing. They would replace the existing 8-micron rated filter cartridges that currently precede the ETF ultraviolet light oxidation (UVOX) treatment unit.

Consideration should be given to installing the ultrafilter with a valving choice, such that it could immediately precede the UVOX unit or follow in the position of the current fine filter. This would allow operators a choice based upon their knowledge of the fouling potential of the aqueous waste to be treated.

Pretreatment should be considered for aqueous wastes such as the N-Reactor fuel basin water. Whether pretreatment constitutes ozonation, iron coprecipitation, coagulation, or ultrafiltration, laboratory and/or pilot-plant tests should be done prior to committing the ETF to accept the aqueous waste for final treatment.

Both lab and pilot-plant scale treatment equipment should be used to differentiate amongst the three preferred ultrafilters for the purpose of treating future aqueous wastes at the ETF. This equipment should also be used to confirm that future aqueous wastes are treatable in the ETF.

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Filtration Engineering Study to Upgrade the ETF

1.0 Scope and Objectives of Study

This study evaluates commercially available filtration technologies which have the potential to augment or upgrade the filtration capabilities of the 200 East Area Effluent Treatment Facility (ETF). Currently, the ETF is scheduled to treat only one waste stream: the condensed overheads from the 242-A Evaporator, referred to as process condensate (PC). When running PC through the ETF, no filtration problems are anticipated, since the ETF was designed to treat PC. However, the ETF will treat other aqueous waste streams in the future which may foul the filtration equipment presently installed in the ETF.

There are two filtration units within the ETF: an 8 micron rated (absolute) filter unit located upstream of the ultraviolet light oxidation (UVOX) unit and a 2 micron rated (absolute) filter unit located upstream of the reverse osmosis unit. Note that an absolute rating generally means all particles at the rated size are removed through filtration, whereas a nominal rating generally indicates most particles at the rated size are removed through filtration. Exact definitions vary from manufacturer to manufacturer.

Figure 1-1 is a flow diagram of the treatment processes and shows the relative orientation of the filters to other units within the ETF. The first filter unit removes the largest particles to protect the (UVOX) unit from scaling and losing organic destruction efficiency by ultraviolet light attenuation. The second filter removes the smallest particles which have a potential to irreversibly foul the reverse osmosis membranes.

Future waste streams are anticipated to contain colloidal and microbiological components. Although the presence of these components has not been confirmed by laboratory analyses, if present, these components would very likely irreversibly foul the filters installed at the ETF. If a broad range of aqueous wastes are to be treated at the ETF, then pretreatment at the generating facility and/or filter upgrades to the ETF would be prudent to ensure successful treatment of these foulants.

The ETF can broaden its acceptance of these waste streams by any of three general types of upgrades: modification of existing equipment, substitution for existing equipment, and/or addition of new equipment. After evaluating applicable filtration technologies, this study explores pretreatment and the three general types of upgrades for broadening waste acceptance. Viable options are formulated and associated costs are estimated. Finally, an implementation plan is proposed involving laboratory and pilot-plant methodologies to ensure future waste streams match ETF filtration waste acceptance criteria.

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Figure 1-1 ETF Process Flow Diagram

2.0 Anticipated Characteristics of Waste Streams to be Treated at the ETF

In addition to the 242-A Evaporator PC, it is anticipated there will be at least five additional kinds of waste streams to treat. Any evaluation of proposed filtration technologies needs to consider the characteristics of these waste streams. Table 2-1 tabulates the waste streams and their associated characteristics. Except for some PC entries, Table 2-1 is only a qualitative summary of what constituents may be present in sufficient concentrations to have a bearing on treatment, including filtration. At present volumetric projections for these waste streams are too uncertain to consider in this study. Never-the-less, forward planning dictates engineers be prepared to treat a hypothetical waste stream that displays a combination of these characteristics. This demands a very robust filtration methodology.

Radioactive Waste: Radioactive components are expected to be found in most of the aqueous wastes under consideration. Accumulation of radioactive waste inventory at the ETF is restricted by its hazard category designation as a "Radiological" facility (WHC 1995a). Because of this restriction, there is no possibility of knowingly receiving aqueous wastes capable of radiolytically damaging any ETF filters postulated for the future. Consequently, radioactive contamination control is the only aspect of ETF treatment that must be accounted for in the ETF filter design.

Regulated Wastes: Many of the aqueous wastes under consideration for treatment at the ETF are considered hazardous and/or dangerous, primarily because they were derived from characteristic wastes or from regulatorily listed sources. Very few (if any) are expected to be characteristically hazardous or dangerous. Those that are characteristic wastes are certainly not expected to be present in high enough concentrations to damage any ETF filters postulated for the future.

Table 2-1 Anticipated Character of Future ETF Aqueous Feeds

	242-A PC	Lab	Fuel Basin	Leachate	Pump & Treat	D & D
Rad	<10 uCi/L	yes	yes	little	little	yes
Hazardous	yes	yes	no	yes	no	yes
Organics	<300 ppm	yes	no	little	some	yes
Ammonia	500 to 1000 ppm	little	no	no	no	no
Heavy Metals	little	some	yes	some	no	yes
Other Inorganics	<100 ppm	some	yes	yes	yes	yes
Colloids	no	no	yes	yes	yes	unknown
Non- Colloidal TSS	little	little	yes	unknown	yes	yes
Micro- biological	little	little	yes	yes	yes	yes

Organics: Many of the aqueous wastes anticipated for treatment in the ETF will contain only trace concentrations of organics (i.e., sparingly soluble at concentrations less than 100 ppm) such that the integrity of any postulated filters will not be affected. However, there remains a possibility that some wastes, especially wastes generated from decontamination and decommissioning (D & D), will contain separable organics and/or emulsions that require pretreatment. Note that this topic is not considered further in this engineering study.

Ammonia: Presently, the 242-A Evaporator PC waste contains ammonia in concentrations that could be as high as 1000 ppm. However, at these concentrations, ammonia is not damaging to any filter postulated for service at the ETF, now or in the future. In addition, there are no known aqueous wastes containing substantially higher concentrations of ammonia that are candidate feeds for the ETF.

Heavy Metals: Heavy metals are not damaging to postulated filter materials. It is not expected they will be present as suspended solids to any appreciable extent. Consequently, based upon their characteristics, they should have no impact on filter selection for the ETF.

Other Inorganics: Inorganics other than heavy metals are also not damaging to postulated filter materials. However, if they are present as suspended solids in sufficient concentrations, they can present a fouling potential that must be taken into account when designing a filtration process.

Colloids: Filters, especially those with absolute ratings of 0.1 to 10 microns, are susceptible to fouling by colloids. Colloids are particles of small diameter (generally in the range of 0.001 to 10 microns) that do not readily settle out of solution but do tend to clump together in a manner that blinds filters. Fuel basin water is suspected of containing colloids. They are probably derived from ferrous and transuranic (TRU) materials that have been and still are present in the basins. When N-Basin water is disturbed, it takes many hours for the water to clear (to the eye), indicating the potential presence of colloids. Another potential source of colloids comes from water pumped out of the ground for subsequent treatment. Such colloids are expected to be siliceous. Iron colloids may be present also. Colloids are a significant concern to industries that need hyperpure water (e.g., the computer chip manufacturing industry). One vendor indicates that siliceous colloids are generally less than 0.1 micron.

Detailed particle size distributions covering the sub-micron range for potential aqueous wastes to be treated by the ETF do not exist. However, it is anticipated that aqueous wastes will contain particles as small as 0.001 microns.

The upper end of the particle size distribution is also not well characterized for most aqueous wastes, but data does exist for the 105 N Fuel Basin. Appendix A contains a particle size distribution for this aqueous waste.

Generally, the distribution extends over a range of 6 to 76 microns. However, the instrument used to measure the distribution appears to have no resolution below 6 microns and it is not designed to measure in the submicron range where colloids exist. According to one vendor, log-normal particle size distributions are often encountered in liquid filtration. Assuming that is the case here, the 105 N Fuel Basin water should contain significant numbers of submicron colloidal particulates.

Microbiological: Aqueous wastes that have sat around for any length of time in contact with ambient air have the potential to grow algae and/or bacteria. Algae and bacteria are carried by the air along with the necessary nutrients for life. Although both have the potential to foul filters, the latter is generally the most problematic. The Savannah River Effluent Treatment Facility had a great deal of fouling problems from a bacteria that was particularly sticky (SRL 1991).

3.0 Available Filters to Choose From

3.1 Filter Characteristics

Filters can be made of multi-component polymers, single-component polymers (i.e., no binders or co-polymers), fibers, metals, and ceramics. Example components include: polypropylene, polysulfone, polyacrylonitrile, polyester, Nylon-66, polyethylene, polyvinylidene di-fluoride, polyfluorotetraethylene, glass fiber, cotton, cellulose, stainless steels (e.g., sintered and woven), and alumina/zirconia ceramic. Appendix B contains a limited chemical compatibility chart for many of these materials.

Filters come in many different configurations, with some being more space efficient. In general the amount of filter surface area obtained per space occupied is in the following order:

Pleated & Spiral String Wound

Spiral Wound Thin-Film Membrane

Hollow Fiber Membrane

1/2-inch Tubular Polymeric Membrane & Ceramic

1-inch Tubular Polymeric Membrane & Sintered or Woven Metal

Flat Sheet.

Other features have little or no impact upon space efficiency but may impact collection efficiency, filter life, strength, and cost. Such features may include:

- o structural webbing for strength,

- o blown polymeric windings, melted together for strength,
- o hydrophobicity to absorb trace amounts of oils,
- o hydrophilicity to shorten the time for a filter to wet,
- o polymeric modifications to induce positive charges which attract negatively charged particulates (most particulates are negatively charged), and
- o bicomponent (polymer within polymer) systems for strength (outer sheaths are melted together), chemical compatibility, and temperature resistance.

3.2 Filter Categories

Vendors of process oriented filters generally consider a microfilter to cover a range of about 0.1 to 10 microns, whereas an ultrafilter is considered to cover a range of 0.001 to 1 micron. There is no distinct break point between the two types of filters. Consequently, this study will consider filters with absolute ratings ≥ 1 micron to be microfilters and filters with absolute ratings < 1 micron to be ultrafilters. The 1 micron breakpoint is convenient for the purposes of this study and does not necessarily reflect any one vendor's approach to particle size ratings.

Note that filters targeted at the pharmaceutical industry define microfilters and ultrafilters differently. These filters usually are rated by Nominal Molecular Weight Cutoff (NMWC). Also, a microfilter used in the pharmaceutical industry would probably be equivalent in micron ratings to an ultrafilter in the waste water treatment industry. This is a consequence of the pharmaceutical industry considering high NMWC membrane filters to be microfilters, whereas, almost all membrane filters are considered to be ultrafilters by the waste water treatment industry.

Figures 3-1 and 3-2 categorize a range of commercially available filters based upon whether they are: microfilters (≥ 1 micron) or ultrafilters (< 1 micron), single use or regenerable, and depth or surface acting. Note that with one exception all of the ultrafilters are surface acting (i.e., collect particles on the filter surface), whereas, all of the microfilters are depth acting (i.e., collect particles throughout the filter thickness).

Figure 3-1. Cartridge Style Microfilters (≥ 1 micron)

Figure 3-2. Ultrafilters (<1 micron)

3.3 Qualified Ultrafilters

This section examines the candidate filters that could potentially be used to upgrade the ETF. Information used to narrow the choices comes from the Savannah River Site, ETF project experience, and filter vendors.

Among the various parameters one must know to choose an appropriate filter are:

- o The particle size distribution in the aqueous waste (Sec. 2.0, Colloids),
- o If the filter materials are compatible with the concentrations of the various constituents in an aqueous waste (Sec 3.1),
- o The capabilities of filters (Sec. 3.1), and
- o How the filter will fit into the overall process (Sec 5.3).

Additional, and perhaps more important parameters include:

- o How the production of secondary waste can be minimized,
- o The fouling potential of the filters,
- o Treatment facility spatial requirements, and
- o Cost.

Whether a particular microfilter or ultrafilter is chosen to augment ETF operations will depend strongly upon two factors: waste minimization and fouling potential. Both factors will influence cost. Waste minimization favors regenerable filters that do not require a pre-coat application. A minimal tendency to foul favors regenerable (i.e., backwashable and washable) filters, but the filters must have a tight enough pore structure to reject particles, especially colloids. Hence, even though some microfilters are backwashable, compressible particles can squeeze through their pores and become irreversibly trapped. Ultrafilters have small pore structures and most are considered to be membranes. Particles do not as readily squeeze into the pores of membranes as easily as they do in other filters.

Depth filters are more likely to irreversibly foul as particles become wedged into their pores. No amount of blowing back with an air or water sparge will dislodge these particles. Whereas, surface filters are made from membranes and typically do not have pores that trap particulates. These filters are regenerated by washing with various chemical agents - often proprietary. Irreversible fouling can occur in these filters when a resistant film builds upon the membrane surfaces. However, these filters take much longer to irreversibly foul than depth filters.

Referring to Figures 3-1 and 3-2, the only filter category that can potentially meet the qualifications are the washable ultrafilters: tubular, hollow fiber, centrifugal, rotating disc, and ceramic. These five ultrafilters are described in Appendix C. The other listed filters require pretreatment to remove colloids (except for the precoat) and to avoid irreversible fouling. A precoat microfilter would have less tendency to foul, but would produce a substantial amount of secondary waste and is not considered further.

With respect to microbiological fouling, vendors tend to recommend hollow fiber and tubular membranes. Not only have these filters been proven in waste water treatment, but there is a successful history of usage in the pharmaceutical/biotechnology and ultrapure water (e.g., for semiconductor chips) manufacturing industries. These industries must contend with algae and bacteria routinely. The Japan Gas Company (ADTECHS 1992) also recognizes that tubular ultrafilters are effective against microbiological fouling. However, even tubular ultrafilters with large lumens can foul. The fouling is a function of having to run the filter tubes in series, with the result the liquid velocity drops off near the end of the filter to make it susceptible to fouling. Sponge balls must be sent down the tube to clean it when this happens.

Ceramic filters may be the worst choice when considering microbiological fouling. Savannah River Site engineers had trouble with microbiological fouling on their ceramic ultrafilters to the extent cleaning was not terribly effective (SRL 1991). It was not until they pretreated their waste water with aluminum nitrate and managed the bacterial count to less than one million bacteria per deciliter that they solved the fouling problem.

When particulate fouling is considered, tubular ultrafilters are considered to be the least problematic, since they can be cleaned with sponge balls. Hollow fiber ultrafilters may be the most susceptible to particulate fouling because of narrower fluid passages, but they are often used by industry when colloidal fouling is a concern. It is not known how resistant centrifugal ultrafilters are to particulate fouling.

With respect to space requirements, the hollow fiber technology is the best, followed closely by the centrifugal technology. The tubular filters would take up the most space. As of this writing, no spatial information was available for the ceramic and rotating disc filters.

Hollow fiber and tubular filters have the longest successful industrial history of the five filters. The centrifugal ultrafilter has a much more limited history, but because it works by creating very strong shear forces at the membrane wall to minimize fouling, it is considered to be a very viable technology. The rotating disc filter would also be considered a viable technology, except that it is too new to have demonstrated its effectiveness in industrial waste water treatment applications.

The cost of a new filter skid to replace the existing ETF fine filter is estimated to be 0.38 to \$1.2 million. Estimates are based upon a rough specification given to filter vendors (as reproduced in Appendix D) and do not include installed costs. The estimates were obtained to gain an idea of what a new skid might cost and how much space they might occupy for the treatment of future aqueous wastes. A byproduct of the exercise was to learn what effect a parameter would have on cost and size. For example, for hollow fiber membranes, cost and size are proportional to the TSS specified for the aqueous waste. Note that the tubular ultrafilters appear to be the least costly followed by the centrifugal ultrafilter, and finally the ceramic ultrafilter. No cost data was available for the rotating disc ultrafilter. However, the number of vendors responding and the rough nature of the specification make this ranking tenuous. Therefore, the existing cost data cannot be used to differentiate between the various filters.

Considering fouling potential, space requirements, and industrial experience, the ceramic and rotating disc filters can be eliminated from further consideration. The former is susceptible to fouling. Whereas, the latter has little or no industrial history. However, it is not easy to rank the other three, since much depends upon the final specifications. The specifications given to the vendors to obtain cost estimates were based upon conservative assumptions about future filtration requirements at the ETF. Too much conservatism (e.g., the 15% TSS specification) could very well have skewed the cost results. Also, configuration requirements (i.e., the spatial orientation) at the ETF were not mentioned in the specifications. These could skew the cost estimates for the filter skids.

4.0 Pretreatment Prior to Filtration

In some cases, it may be prudent to pretreat a waste stream prior to filtration at the generator's facility. The reasons for doing so would be related to the contaminant loading of the waste stream, where contaminants include: TSS, TDS, organics, and radionuclides. If the concentration of these contaminants exceed the ability of the ETF to treat them, then pretreatment may be necessary. For example, the ETF is designated as a radiological facility and cannot accumulate more than a certain amount of each radionuclide (WHC 1995a).

There are several methods to accomplish pretreatment. Four potentially promising ones are addressed in this section. The four are:

- o Ozonation to oxidize iron(II) to iron(III),
- o Coprecipitation,
- o Coagulation with aluminum hydroxide, and
- o Ultrafiltration.

4.1 Ozonation

Ozonation of aqueous wastes is an attractive pretreatment, if colloidal iron and/or manganese are present. It has an additional benefit of degrading most organics (if present) to smaller molecular weight compounds. The process may be useful in treating fuel basin water, which is believed to contain colloidal iron and heavy metals. Colloidal iron exists as Fe(II) and when ozonated forms Fe(III) in the form of an iron hydroxide. Iron hydroxide when formed will precipitate out of solution and adsorb heavy metals. This process is known as coprecipitation. Coprecipitation will only be significant, if the amount of iron is present in sufficient concentration. What constitutes a significant concentration can only be determined through testing. As a reference point, the 300 Area Treated Effluent Disposal Facility uses 20 ppm iron in its co-precipitation process.

4.2 Iron Coprecipitation

Iron coprecipitation has been proposed for treating groundwater and various other liquid effluents at Hanford (WHC 1988). As stated above, the process is a combination of precipitation and adsorption. The first step in coprecipitation is a pH adjustment to between 2 and 4 which conditions the aqueous waste for the next step. The next step is to add ferrous sulfate at about 0.2 lb of iron per 1000 gallons (or equivalently, 65 mg/L of FeSO_4) which will dissolve and disperse into solution at the pH established in the first step. Hydrated lime is then added to raise the pH and precipitate iron as the oxyhydroxide, $\text{FeO}(\text{OH})$. Heavy metals can either adsorb onto the oxyhydroxide or coprecipitate as metal hydroxides. Precipitated materials are then removed by passing the heterogeneous solution through a 0.1 micron (nominal) tubular membrane microfilter made of polyfluorotetraethylene.

Tests on groundwaters showed secondary waste production amounted to 0.4 to 5.8 gallons per 1000 gallons treated at a dry solids content of 20% to 52% (or equivalently, 210 to 1200 mg per liter treated, assuming completely dried waste has a density of 1 g/cc). Tests on various other aqueous wastes showed coprecipitation to be promising in removing most heavy metals. However, membrane fouling (thought to be caused by silicates) was a problem in treating laundry waste waters by coprecipitation/filtration.

4.3 Coagulation

Coagulation with aluminum compounds has been tried by Savannah River Site (SRS) personnel (SRL 1991) and the French (Lahoussine-Turcaud 1990 and Pouet 1994). Two different methodologies were applied: direct addition of aluminum compounds and electrocoagulation.

Both coagulation methods were tried by personnel at the SRS waste water treatment facility, a facility somewhat similar to the 200 Area ETF. The SRS facility utilizes pH adjustment, microfiltration (filter made of alpha alumina ceramic), and reverse osmosis. Both biofouling and inorganic fouling of the SRS microfilter (nominal rating 0.2 microns) occurred to the extent that

throughput was severely diminished (from 300 gpm to 60 gpm). Biofouling was attributed to bacteria with very sticky exopolysaccharides. Inorganic fouling was attributed to colloids consisting of aluminum, silica, and iron. An attempt was made to electrocoagulate both types of foulants. A vendor's aluminum electrocoagulator (probably a single electrode), generated unsatisfactory results. Direct addition of aluminum nitrate worked best. Tests showed the optimum formula was to add 67 to 135 mg/L of Al(III) (equivalently, 530 to 1100 mg/L as $\text{Al}(\text{NO}_3)_3$) at a pH below 2.7, followed by neutralization to a pH between 6.5 and 8.5. It is believed the resulting aluminum hydroxide formed a polymer with a spinel structure. The polymer trapped bacteria and colloids (probably through electrostatic attraction) to form larger particles which were more easily filtered.

The French (Lahoussine-Turcaud 1990) tried filtering river water from the Seine using a hollow fiber membrane filter (rated at 0.001 micron) made of polysulfone. Significant irreversible fouling occurred, even if the water was first subjected to coagulation by the addition of a commercially available polyaluminum product. Similar to the SRS situation, it was suspected that microbial polysaccharides were responsible for the fouling.

Another French source (Pouet 1994) tried pretreating municipal waste water with an electrocoagulator followed by a 0.1 micron rated ceramic microfilter. The electrocoagulator consisted of charged (direct current) aluminum plates emersed in waste water within a vessel. When driven to the optimum current density, gas evolved from the plates to float undissolved solids to the top of the vessel where they could be skimmed off. Aluminum hydroxide flocculent was formed from the electrode plates to capture colloidal materials. The results were less than satisfactory, but irreversible fouling was reduced from the case where only a microfilter was employed.

4.4 Ultrafiltration

Ultrafiltration as described above in Section 3.3 can be used to pretreat aqueous wastes at the generator. It is an effective method for removing colloidal materials but produces a concentrate stream (a secondary waste) that must be treated further (e.g., via evaporation). In addition, a rough filter is generally required to protect an ultrafilter from plugging. Consequently, purchasing a roughing filter and an ultrafilter for every aqueous waste that needs pretreatment could become expensive, unless the pretreatment skid could be used over and over again. In addition, the pretreatment skid would need to be sized to treat the largest aqueous waste stream, unless that stream had its own dedicated pretreatment system. Ultimately, if enough streams needed individual pretreatment, it would become cost effective to only provide such treatment at the ETF.

4.5 Pretreatment Summary

Considering the four pretreatment methods discussed in this section, ozonation produces the least amount of secondary waste. However, ozonation only works if sufficient iron is already present. Both coprecipitation and coagulation

require chemical additions be made to remove colloidal iron. Only ultrafiltration and coagulation by addition of aluminum nitrate appear to be successful in removing colloidal silica. Coprecipitation and coagulation additions result in higher secondary waste production than expected from ozonation. However, applied to Hanford aqueous wastes, all four methods would probably require about the same amount of equipment, including membrane separators and settling vessels.

5.0. Options to Broaden ETF Waste Acceptance

As stated in Section 1, there are three filtration-related upgrades that can be used to broaden the acceptance of aqueous waste for treatment at the ETF. They are: modification of existing ETF equipment, substitution of new equipment for existing equipment, and addition of new equipment. When pretreatment at the generating facilities is included, there are four methods that can be used to improve the overall treatment of aqueous wastes.

5.1 Pretreatment

As discussed in Section 4, four viable options involve ozonation, coprecipitation, coagulation, and ultrafiltration. For fuel storage basin water, ozonation appears to be the best option. Ozonation has the potential to remove colloidal material consisting of heavy metals, including radioactive species. Hence, ozonation can reduce the fouling potential at the ETF, help keep the inventory of radioactive material below ETF hazard category limits, and help keep the ETF from generating secondary waste that exceeds TRU concentrations.

However, without further laboratory and/or pilot-plant testing, it is not known how effective ozonation can be in removing colloids from fuel basin water. Nor is it known what the best filter would be for removing precipitates from the ozonated water. If ozonation proves not to be the best option for removing colloids, then ultrafiltration, coprecipitation and coagulation could be considered.

For groundwaters containing siliceous colloids, only coagulation and ultrafiltration have a reasonable chance of being effective. The degree of efficacy is unknown without testing. SRS personnel found coagulation effective, but the SRS waste water contained iron and silica colloids, bacteria, and algae. Such a matrix is likely to be different from Hanford groundwater. It is also not conclusive if Hanford groundwater would need to be pretreated before final treatment at the ETF. Coagulants could be added at the ETF instead of pretreating at the generation point. However, it would probably be best to apply ultrafiltration at the generation point with subsequent treatment of the concentrate at the ETF. Further analysis is needed to be sure.

5.2 Modification of Existing Equipment

None of the three recommended filters described in Section 3 can be retrofitted to the spiral (string) wound polypropylene cartridge filters that currently exist in the ETF. These technologies would need to replace them or be added to existing the equipment, if they were to be incorporated into the ETF.

5.3 Substitution for Existing Equipment

As stated in Section 1.0, there are two filtration units within the ETF: an 8 micron rated (absolute) filter unit and a 2 micron rated (absolute) filter unit. These are referred to as the rough and fine filters, respectively. The first filter is located upstream of the UVOX units and is meant to remove the largest particles from aqueous wastes. If these particles were not removed, then the ultraviolet light in the UVOX unit would be attenuated and organic destruction would be less efficient. The second filter is located upstream of the reverse osmosis units to protect the same from irreversible fouling. As discussed in Section 3, these two filters are of a rating that would tend to foul irreversibly from colloids and bacteria.

It would be best to substitute one of the top ranked filters (hollow fiber membrane, tubular ultrafilter or centrifugal ultrafilter) for the existing fine filtration unit. Because the rough filter can be a cartridge type backflushable filter, the existing unit's housing could be utilized. The rough filter would need to be rated between 100 and 400 microns to protect the fine filter's liquid passages from plugging with large particles, while allowing bacteria and colloids to pass without plugging the rough filter. Plugging the fine filter with large particles would especially be a concern for hollow fiber membrane filters which generally cannot handle more than 15 weight% total suspended solids. Note that recommended lumen size for hollow fiber filters is on the order of 0.5 to 2.5 mm.

If the above substitution takes place, then consideration must be given to what effect it has on the treatment efficiency of the UVOX unit. In particular, if colloids and bacteria are allowed to pass into the UVOX unit, at some threshold concentration they would begin to attenuate the ultraviolet light. A lowered organic destruction efficiency would result. To solve this, the fine filter could be placed immediately in front of the UVOX unit. However, if the organic constituents were incompatible with the filter membrane, then it would be best to adopt the current process flow in which the fine filter is downstream of the UVOX unit. A compromise would be to allow for either configuration through appropriate valving.

5.4 Addition of New Equipment

The addition of new equipment to the ETF has the potential to be problematic from an operational and regulatory stand point. Assuming there is room, adding equipment without removing the old may complicate the running of the

facility unnecessarily. Furthermore, regulators might determine that the addition of new equipment would change the basis upon which they originally approved the ETF as best available technology. However, these are not likely to be insurmountable problems.

One potentially viable approach would be to provide an optional ultrafilter that could be valved into the process whenever engineers determine the existing system is likely to irreversibly foul. In fact, if a determination is ever made that future hard-to-filter aqueous wastes will be limited in volume, then the ultrafilter could be rated at a fraction of the existing ETF capacity. Note that the minimum ETF capacity is 40 gpm. This means the ultrafilter capacity (because of internal recycle) would need to be between 45 and 50 gpm.

6.0 Permitting Impacts

Once the Resource Conservation and Recovery Act (RCRA) permit for the ETF is issued, changes to the ETF filtration system will trigger additional permitting activities. If the ETF filters are modified or replaced, then a Class I change to the RCRA Part B permit will be required. Such a Class I change would probably not require the written approval of Ecology per WAC 173-303-830. If additional capability is engineered into the ETF treatment train, then a more involved change would likely be required - a Class II change. However, formalized changes will not be required until the RCRA Part B permit is issued. Issuance is likely to occur after any filtration upgrades are made. Consequently, the draft Part B permit would be updated as details of the filtration upgrade become known.

7.0 Acceptance Testing of Aqueous Wastes for Treatment at the ETF

7.1 Waste Acceptance Process

The treatment of hazardous and dangerous aqueous wastes at the ETF is a regulated activity. Only regulated wastes allowed by the RCRA Part A permit and those wastes which are not designated hazardous or dangerous can be treated at the ETF. Regardless of the designation, aqueous wastes must be characterized prior to treatment (with the exception of PC due to prior characterization data). Regulated wastes are required to be characterized (WHC 1995b), but unregulated wastes may need to be characterized if engineers believe treatment of the aqueous wastes might be problematic. Laboratory and/or pilot-plant tests will then be necessary to assure treatability. If engineers believe treatment is not feasible at the ETF, the generator will need to pretreat the waste (e.g., as discussed previously in Section 4).

7.2 Acceptance Testing

Pilot-plant scale testing under an RD&D permit has been performed with an ETF style rough filter on process condensate at the Liquid Effluent Retention Facility (LERF). However, approaches to laboratory and/or pilot-plant acceptance testing for treatment of aqueous wastes at the ETF needs to be

developed, since no filter testing has been done with other potential ETF feeds. It would be advantageous to be able to anticipate waste water/filter interactions in advance of requests to treat these feeds. This is true whether or not the filters at the ETF are upgraded per the recommendations of this study. Therefore, laboratory and pilot-plant filtration equipment should be made available to collect data on capacity, inorganic and microbiological fouling, and cleaning methodologies.

7.3 Capacity Tests

Capacity tests can determine maximum expected flow rates for a given filter system size. These tests can also provide information on how often the filters will need to be backwashed or cleaned in place. Because (according to vendors) these once-through tests must be done at minimum flow rates of between 1 and 5 gpm and run a minimum of 4 to 48 hours, they are beyond laboratory scale. Exceeding laboratory scale flow rates is problematic if the aqueous wastes are regulated. Specifically, treatment of more than 1000 Kg of wastes regulated by the Resource Conservation and Recovery Act (RCRA) require an RD&D permit. These permits can take up to two years to put in place. Except for the RD&D permit currently in force to treat PC, it is not practical to test under these permits. Therefore, for capacity tests on regulated wastes, engineers may be relegated to obtaining data through the use of synthetic aqueous wastes and/or rely on laboratory tests. The former is a poor substitute for the real thing and the latter can be problematic to scale up.

7.4 Inorganic and Microbiological Fouling Tests

Definitive inorganic and microbiological fouling tests are difficult to design. The degree of difficulty is somewhat dependent upon the type of aqueous waste, whether or not it is regulated or radiological. These factors determine where the tests can be performed and with how much inventory. Other factors involve the limitless microbiological diversity, the complicated interactions between bacterial and inorganic foulants, and interactions between particles and filter materials. Accounting for these factors would require a huge number of tests. Since performing a huge number of tests is not practical, engineers will need to rely upon a lesser amount of tests intended to qualitatively measure the effects of these factors.

To the extent practical, tests on relatively small amounts (i.e., under 1000 Kg) of regulated and radioactive wastes must be performed in laboratory facilities set up physically and administratively to handle small quantities of mixed waste. Tests on non-regulated and non-radiological wastes can be performed at any laboratory, pilot plant, or generator's facility without regard to inventory restrictions.

Tests on small volumes of waste are envisioned to utilize vendor supplied bench-top scale filtration units. After running waste through the bench-top units, the filter cartridges could be cut open and coupons obtained to characterize filtered microorganisms. The microorganisms could then be

categorized by their metabolic characteristics (e.g., heterotrophic). Microscopy could be used to evaluate the numbers and morphology of microorganisms and whether they are attached to inorganic particles.

7.5 Cleaning Tests

Cleaning agents could be applied to filter coupons loaded with bacteria and inorganic foulants to determine which agents clean best. Coupons could be cut from laboratory scale filters or could be made of single-pass flat sheet filters that are normally used by the pharmaceutical industry. Appendix E lists several potential cleaning agents. Proprietary agents are also available from filter vendors. Tests could also be designed to challenge laboratory-scale filters with silica- or iron-based colloids from actual wastes or from simulated matrices. Engineers could look for throughput problems after backwashing the filters. From these tests, clean-in-place methods could then be recommended for full-scale filters.

7.6 Recommended Test Equipment

To perform the above tests in Section 7, both pilot-plant scale (less than 25 gpm) and laboratory (less than 1 gpm) equipment will be needed. As available, the top three rated ultrafilters should be represented amongst this equipment. Laboratory scale is appropriate for qualitative studies. Whereas, pilot-scale is appropriate for quantitative studies.

The Chemical Engineering Laboratory already has a centrifugal ultrafilter rated at 5 to 10 gpm and a tubular ultrafilter rated at 5 gpm (but upgradable to 25 gpm). The tubular ultrafilter equipment can be reconfigured to house hollow fiber cartridges. Therefore, hollow fiber membrane cartridges should be purchased to retrofit the existing equipment so that a comparison between the three different filter technologies can be conducted. Data obtained from such a test would also aid designers in scaling up to a full size filtration system.

Virtually all filter vendors offer pilot-plant scale equipment, but not all offer laboratory scale equipment. Centrifugal and tubular ultrafilters are not readily available at laboratory scale. Fortunately, hollow fiber membrane filters can be purchased at this scale. Laboratory scale is appropriate for analyzing the types of foulants that might collect on filter surfaces. This scale is also appropriate for determining which cleaning agents can remove foulants.

Characterization of foulants and identification of the best cleaning agents may be appropriate tools in accepting aqueous waste for treatment in the ETF. This is especially true if engineers believe an aqueous waste contains colloids. The presence of colloids is usually predicted by determining solution chemistry and measuring the particle size distribution. Determining the particle size distribution is not easy in the submicron range (Nicoli 1995). Such equipment may need to be purchased if laboratory support is not available.

8.0 Recommendations

To ensure the ETF will be able to treat these future waste streams successfully, the existing ETF fine filter (i.e., the 2-micron rated microfilter) should be replaced with one of three ultrafilters (hollow fiber, tubular, or centrifugal). Such a filter should not foul in the presence of colloids and microorganisms.

Alternatively, a decision could be made to add ultrafilter capacity to the existing ETF filters. Problematic aqueous wastes could be treated by valving in the standby filters. If the amount of wastes needing treatment by ultrafiltration were limited (and this has yet to be determined), then the capacity of the ultrafilter could be less than the rated capacity of the ETF.

To protect the ultrafilter, rough filter cartridges rated at between 100 and 400 microns should function as prefilters. The new rough filter cartridges could be installed into the existing rough filter housing. They would replace the existing 8-micron rated filter cartridges that currently precede the ETF ultraviolet light oxidation treatment unit.

Consideration should be given to installing the ultrafilter with a valving choice, such that it could immediately precede the UVOX unit or follow in the position of the current fine filter. This would allow operators a choice based upon their knowledge of the fouling potential of the aqueous waste to be treated.

Pretreatment should be considered for aqueous wastes such as the N-Reactor fuel basin water and groundwater obtained from pump-and-treat operations. Whether pretreatment constitutes ozonation, iron coprecipitation, coagulation, or ultrafiltration, laboratory and/or pilot-plant tests should be done prior to committing the ETF to accept the aqueous waste for final treatment.

Both lab and pilot-plant scale treatment equipment should be procured by the Chemical Engineering Laboratory to differentiate amongst the three preferred ultrafilters for the purpose of treating future aqueous wastes at the ETF. This equipment combined with existing pilot-plant equipment should also be used to confirm that future aqueous wastes are treatable in the ETF. Consideration should be given to purchasing a sub-micron particle size analyzer to determine the presence of colloids.

9.0 References

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5. (Lahoussine-Turcaud 1990) Lahoussine-Turcaud, V., Wiesner, M.R., Bottero, J., and Mallevialle, J., "Coagulation Pretreatment for Ultrafiltration of a Surface Water," Research and Technology, December 1990, pp 76 - 81.
6. (Pouet 1994) Pouet, M.-F. and Grasmick, A., "Electrocoagulation and Flotation: Applications in Crossflow Microfiltration," Filtration and Separation, May 1994, pp 269 - 272.
7. (Nicoli 1995) Nicoli, D.F., Wu, J.S., Chang, U.J., McKenzie, D.C., and Hasapidis, K., "Wide Dynamic Range Particle Size Analysis by DLS-SPOS, A Combination of Two Technologies," American Laboratory, April 1995, pp 41-49.
8. (WHC 1995b), *Waste Analysis Plan for the 200 Area Effluent Treatment Facility and the Liquid Effluent Retention Facility*, WHC-SD-EN-WAP-001, Rev. 0, September 1995.

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Appendix A Chemical Compatibility Table for Selected Filter Materials

Chemical	Polysulfone	Polyethylene
Acids		
acetic acid, < 5%	acceptable	acceptable
acetic acid, > 5%	short term only	acceptable
citric acid	OK for < or = 1%	no data
formic acid	OK for < or = 1%	no data
glacial acetic acid	no data	acceptable
HCl	OK for < or = 0.01 N	OK for concentrated
nitric acid	short term for < or = 1%	OK for concentrated
phosphoric acid	OK for < or = 0.1 N	acceptable
oxalic acid	OK for < or = 1%	no data
sulfuric acid	OK for < or = 1%	OK for concentrated
chromic acid, conc.	no data	acceptable
Bases		
ammonium hydroxide	OK for < 5%	OK for 6 N
KOH	no data	OK for 3 N
NaOH	OK for < or = 1 N	OK for 6 N
Alcohols		
butanol	OK for < 1 %	acceptable
ethanol	OK for < or = 10 %	acceptable
methanol	OK for < or = 10 %	acceptable
Ketones		
acetone	no data	acceptable
methyl ethyl ketone	not recommended	acceptable
methy isobutyl ketone	no data	acceptable
Halogenated Hydrocarbons		
carbon tetrachloride	no data	short term only

Chemical	Polysulfone	Polyethylene
chloroform	not recommended	not recommended
methylene chloride	not recommended	short term only
Aromatic Hydrocarbons		
benzene	no data	not recommended
toluene	not recommended	not recommended
xylene	not recommended	not recommended
Others		
amines	not recommended	no data
dimethyl formamide	not recommended	acceptable
formaldehyde	OK for < or = 1%	OK for 4%, short term for 37%
glycerine	OK for < or = 2%	acceptable
hydrogen peroxide	short term for < or = 1%	no data
sodium hypochlorite	OK for < or = 300 ppm short term for > 300 ppm	no data
sodium nitrate	OK for < or = 1%	no data

Notes:

Data are taken from vendor literature. Additional information can be found in the Cole-Parmer Instrument Company catalogue, Toll-free number (800) 323-4340.

"OK" means that the solutions are compatible.

"Not Recommended" generally applies to all concentrations. However, for guidance on low ppm concentrations and lower, the filter vendor should be consulted.

"Short-Term" is open to interpretation. For specific recommendations, the vendor should be consulted.

Appendix B N-Reactor-Fuel-Basin Water Particle Size Distribution

Figure 1. 105 N Fuel Basin Solids Particle Size Distribution.

Particle Size Distribution

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Appendix C Descriptions of Selected Filter Technologies

Centrifugal and Tubular Polymeric Ultrafilters (as excerpted from C.V. King to J.W. Kelly, Internal Memo 28130-91-081), 8/7/91)

Centrifugal ultrafiltration is made up of flat membrane disk packs stacked in a horizontal row. The influent is fed into the hollow center while the filter pack rotates. After passing through the face of the filter medium the filtrate is expelled through circumferential holes in the polyethylene terephthalate retainer rings. The concentrate, which is unable to pass through the thin film filter membrane made of polyvinylidene fluoride, passes through the outer holes of the filter membrane packs and is eventually discharged. The outer holes are sealed to prevent mixing of the filtrate with the concentrate. The driving force of the expelled concentrate is a differential pressure of 150 psig. The housing is held together with a steel clamping and O-ring arrangement where the influent is fed through one drive shaft and the concentrate is discharged through the opposite drive shaft.

Tubular ultrafiltration is a low pressure (10-150 psig) membrane process for separating suspended solids and high molecular weight dissolved materials from liquids. Fluid flows across the membrane surface at high velocity. This cross-flow characteristic differs from the perpendicular flow of ordinary filtration, where a "cake" builds up on the filter surface requiring frequent filter replacement or cleaning. If bacteria or colloids build up on the membrane surface a spongeball can be fed through the filter tube during operation to remove the contaminants from the membrane.

Hollow Fiber Membrane Filters

Hollow fiber membrane filters are available as cartridge units. They consist of many tubules running parallel within a plastic housing. Both ends of the housing are potted with a polymer. In operation, the feed stream flows through the inside (lumen) of the membrane channels (in a single pass) until it becomes concentrated to become the reject stream. Permeate passes through the membrane walls and is collected on the shell-side of the cartridge.

Ceramic Membrane Filters

Ceramic membranes consist of an engineered cylindrical monolith. Each monolith contains a large number of passageways which extend from an inlet end face to an outlet end face. The monolith is modified by converting some of the passageways to filtrate conduits. After making this modification, which eliminates the flow-limiting resistance of the porous support, ceramic membrane coatings are applied to the remaining monolith passageways. One or more membrane layers are applied and sintered to form a stable, strongly bonded ceramic membrane. Materials used for the different membrane layers include alumina, zirconia, or other proprietary materials.

During operation, a pressurized liquid feed is introduced at the inlet end face of the unit and concentrate is removed at the opposite end face. Filtrate which passes through the membrane coating enters the monolith and flows through the filtrate conduits to a filtrate collection zone located along the side of the unit.

Rotating Disc Membrane Filter

The rotating disc membrane filter is a relatively new product initially targeted at the biopharmaceutical industry. The vendor claims it takes up less space than other filters with the same membrane area. It is designed to filter aqueous streams that would otherwise foul conventional filters. This is accomplished by the creation of shear forces at the membrane surface. Contaminants are lifted away before small particles can penetrate the membrane surface and cause irreversible fouling. Shear is created in a gap between a rotating solid disc and the membrane surface. While concentration polarization occurs to a degree, the shear controls it and prevents the buildup of a gel layer. Note that gel layer buildup typically occurs whenever microbiologic materials are present. Gel layer formation is for the most part reversible through flushing. However, gel layer buildup is not desirable since it increases membrane permeation resistance.

Appendix D Rough Cost Estimates from Vendors

Given to vendors:

Currently, our facility has a 2 micron (absolute) filter to protect reverse osmosis membrane filters. We are concerned that bacterial and colloidal matter (Fe & Si) may be present in waste waters we plan to treat in the future; hence the reason for the study.

Given a generic specification for waste water treatment, can you give me a rough estimate of:

- o How much hollow fiber membrane and tubular membrane ultrafilter systems would cost to purchase,
- o How much they would cost to operate, and
- o What the footprint would be for each?

Generic Specifications

Flow Rate: 40 - 170 gpm; the system is to be composed of internally redundant components (e.g., three 60 gpm subunits, so that we can run at 2/3 of flow rate while changing out 1/3)

Campaign Time: Assume the facility can operate for 40 days, 24 hours per day at which time maintenance is done.

Instrumentation: Monitor flowrate, temperature, turbidity, and pressure drop only across each filter unit through a distributed control system.

Particle size distribution: 0.001 to 200 um; assume a 200 um prefilter is in front of the ultrafilter.

Organics: Assume the organic loading is only 1 -2 ppm.

TDS: 5,000 ppm, includes 2,000 ppm ammonium sulfate.

TSS: 1-15% by weight.

Other: pH 4 - 6, temperature 35 ° C.

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Rough Cost Estimates Received (Millions of Dollars):

<u>Vendor</u>	<u>Centrifugal UF</u>	<u>Ceramic UF</u>	<u>Tubular UF</u>	<u>Hollow Fiber</u>	<u>Notes</u>
A	0.5 - 0.6		0.4 - 0.5		1
B		1.0 - 1.2			2
C				0.72 - 0.80	3
D			0.38		4

¹Each system is designed to have three filters on-line and one off. Each centrifugal filter unit has 140 ft² of membrane area. The centrifugal unit membrane is PVDF with 100 K molecular weight cutoff. The footprint of the centrifugal filter is 300 ft² and the tubular filter footprint is 700 ft².

²The ceramic ultrafilter is prone to fouling by colloids.

³The cost estimate was based on a very conservative model in which an ultrafilter was used to clean up the ash pond from a coal fired power plant. Therefore, a 15% solids loading was assumed. If the solids loading is cut in half, then the price is cut approximately in half. The design uses three 50 gpm skids (50 gal/ft²/day). Each skid has fifteen 100-square foot hollow fiber cartridges, a 40 HP, 2000 gpm recirculation pump, a backwash tank, and a 20 HP backwash pump. Each skid has a footprint of 70 ft², for a total footprint of 210 ft².

⁴Three filter units are on-line and one is off. The four units have a total of 8,294 ft² in membrane area. Each unit contains 288 ten-foot tubular membranes with polyvinyl chloride (PVC) or CPVC plastic shells manifolded into 24 parallel passes of 12 tubes in series per pass. Overall dimensions for each unit are 11.7 ft. long by 8 ft. wide by 9.25 ft. high, a footprint of 375 ft². Ten feet of clearance must be allowed at one end of each unit for membrane removal.

Appendix E Commonly Used Cleaning Agents for Polymeric Filters

<u>Cleaning Agents</u>	<u>Notes</u>
citric acid	removes iron complexes
alkaline cleaner	removes iron complexes
nitric acid	removes mineral scale
phosphoric acid	removes mineral scale
oxalic acid	no information
hydrochloric acid	no information
acetic acid	no information
Notes:	<ol style="list-style-type: none"> 1) Chlorides (from HCl) are not compatible with ETF equipment. 2) Cleaning agents are different from regenerants which are added to the surface of a membrane filter. They form temporary, but relatively long-lasting films that act as membranes to protect the filter membrane from fouling.

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