APPLICATION OF ARCHIMEDES FILTER FOR REDUCTION OF HANFORD HLW

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ABSTRACT

Archimedes Technology Group, Inc., is developing a plasma mass separator called the Archimedes Filter that separates waste oxide mixtures ion by ion into two mass groups: light and heavy. For the first time, it is feasible to separate large amounts of material atom by atom in a single pass device. Although vacuum ion based electromagnetic separations have been around for many decades, they have traditionally depended on ion beam manipulation. Neutral plasma devices, on the other hand, are much easier, less costly, and permit several orders of magnitude greater throughput. The Filter has many potential applications in areas where separation of species is otherwise difficult or expensive.

In particular, radioactive waste sludges at Hanford have been a particularly difficult issue for pretreatment and immobilization. Over 75% of Hanford HLW oxide mass (excluding water, carbon, and nitrogen) has mass less than 59 g/mol. On the other hand, 99.9% of radionuclide activity has mass greater than 89 g/mol. Therefore, Filter mass separation tuned to this cutoff would have a dramatic effect on the amount of IHLW produced…in fact IHLW would be reduced by a factor of at least four.

The Archimedes Filter is a brand new tool for the separations specialist's toolbox. In this paper, we show results that describe the extent to which the Filter separates ionized material. Such results provide estimates for the potential advantages of Filter tunability, both in cutoff mass (electric and magnetic fields) and in degree of ionization (plasma power).

Archimedes in now engaged in design and fabrication of its Demonstration Filter separator and intends on performing a full-scale treatment of Hanford high-level waste surrogates. The status of the Demo project will be described.

INTRODUCTION

The Archimedes Filter represents a next generation advance in separations technology. Never before has it been practical to separate large amounts of material atom by atom in a single pass device. Although vacuum ion based electromagnetic separations have been around for many decades, they have traditionally depended on ion beam manipulation. Ion beam separations are exemplified by so-called Calutrons, which were developed as a result of the Manhattan project to separate uranium and other isotopes from a mixture of isotopes.

Calutrons suffer from very high cost and very low throughput. These limitations are associated with the difficulty of creating and manipulating pure ion beams in a vacuum. Neutral plasmas, on the other hand, are much easier and less costly to create and permit several orders of magnitude greater throughput. On the other hand, the ways that individual ions are manipulated in neutral plasmas are more limited than with ion beams. Nevertheless, the Archimedes Filter represents a straightforward application of plasma principles to effect a separation into two fractions: a heavy and a light. Although the mass width of the Filter around the cutoff mass is around 20% of the mass, the separation outside of the mass cutoff range is, in principle, perfect.

The Filter operates in a manner that is similar but distinct from a plasma centrifuge. While the plasma centrifuge is highly collisional and represents a continuous incremental mass separation, the Filter condition is collisionless and its mass separation is discontinuous. Above the cutoff mass, ions are not confined and below the cutoff mass they are confined. In addition, the stability of ion orbits is independent of ion origin.

Therefore the Filter has many potential applications in areas where separation of species is otherwise difficult or expensive. In particular, radioactive waste sludges at Hanford have been a particularly difficult issue for pretreatment and immobilization. Unlike the waste sludges now being vitrified at Savannah River Site's Defense

Waste Processing Facility (SRS DWPF), Hanford's waste sludges are much more complex. Therefore many aspects of particular Hanford waste sludges will necessarily complicate those sludges' vitrification. For example, bismuth, phosphate, and uranium are at very high concentrations in bismuth phosphate waste and correspondingly lower concentrations in other sludges. Another example is that chromium and Boehmite are present in Redox waste but virtually absent in all other sludges.

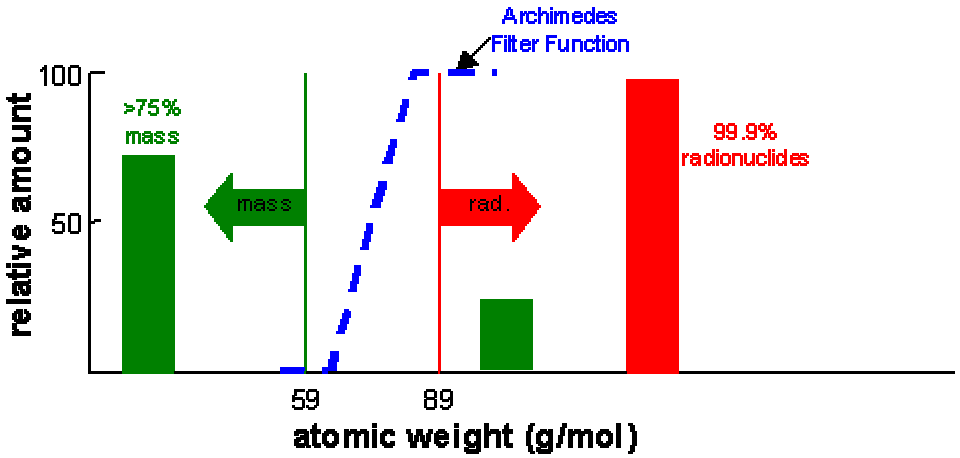

Fig. 1. Archimedes Filter function in "gap" between Hanford high level waste sludge mass and mass of 99.9% of radionuclides.

Figure 1 shows the Archimedes Filter function for plasma with ions in the +1 state and applies to Hanford HLW. Here over 75% of the sludge (excluding water, carbon, and nitrogen) has mass less than 59 g/mol while 99.9% of radionuclide activity has mass greater than 89 g/mol. Therefore, Filter mass separation tuned to this cutoff would have a dramatic effect on the amount of IHLW produced…it would be reduced by a factor of four.

The magnetic and electric fields determine Filter position according to the relation

$$A_{cutoff} = \frac{m_c}{Zm_H} = \frac{B^2 R_{wall}^2}{8V}\frac{e}{m_H} \qquad \text{(Eq. 1)}$$

For example, for:

B = 1500 gauss
V = 530 volts
R_{wall} = 0.40 m

then

A_{cutoff} = 84 g/mol (cut-off atomic weight).

A_{cutoff}	atomic weight of atom at cutoff
m_c	mass of cutoff atom
m_H	mass of hydrogen
Z	ion charge
B	magnetic field
R_{wall}	plasma radius
V	center to wall voltage
e	electronic charge
m_H	mass of hydrogen.

Plasma geometry and density distribution determine the Filter function slope or width.

APPLICATION TO HANFORD WASTE REDUCTION

For application to Hanford, the most effective use of the Filter would be for reduction of oxides that are bound for the HLW melter. Although other applications for decontamination of the LAW stream are certainly possible, this paper will focus on the scheme shown in Fig. 2.

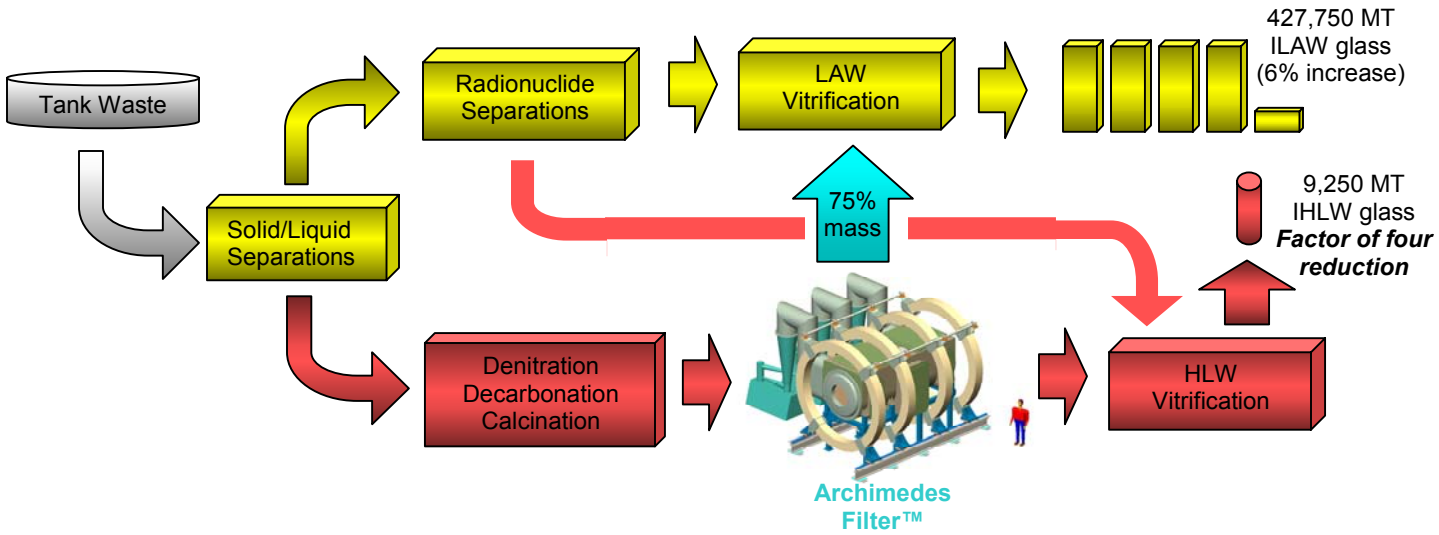

Fig. 2. Schematic showing placement of Filter in HLW oxide stream.

The solids/liquids split shown here is that from Kirkbride, et al., where 95% of the sodium is routed to the LAW stream as well as 80% of the aluminum. We apply the filter to the HLW branch following solids/liquid separation and after a denitration, decarbonation, and calcination step, subsequently sending 75% (or more) of the HLW oxides back to the LAW stream. Although this results in a 6% increase in the ILAW product, a factor of four or more reduction in HLW glass suggests significant overall cost reduction.

This factor of four reduction comes from removal of elements lighter than mass 84 from the HLW feedstock. Radionuclides in the waste are largely in the heavy mass fraction and Table I shows decontamination factors for selected radionuclides.

Table I. Decontamination factors predicted for selected radionuclides.

Radionuclide	Decontamination Factor
Sr-90	185
Tc-99	>1000
Cs-137	>1000
TRU	>1000

Note that Sr-90 is close to the Filter mass cut-off and therefore is not completely removed from the light fraction while the heavier radionuclides are far from the Filter mass cut-off and therefore are in principle completely removed from the light fraction. There will undoubtedly be some practical limits for these decontamination factors and we expect to measure those decontamination factors in our demonstration program that is described below.

FILTER SYSTEM

The Filter system consists of four primary subsystems:

1. Feed preparation;
2. Injection and vaporization;

3. Ionization and separation;
4. Collection of light and heavy oxides.

Feed preparation

Given the large amount of sodium present in the Hanford waste along with the desire to minimize the atom-mols of material to process in the Filter, a natural "solvent" for Hanford high level waste is molten sodium hydroxide. Following denitration, decarbonation, and calcination sodium hydroxide is by far the largest component in Hanford waste as shown in Table II [from Kirkbride, el al. 1999].

Table II. Weight percent oxides in Hanford HLW blend.

Oxide	**Weight percent**	**Weight percent with increased NaOH**
NaOH	36%	50%
Fe2O3	12%	9.6%
Al2O3	12%	9.3%
SiO2	7.2%	5.6%
P2O5	3.0%	2.4%
CaO	2.6%	2.0%
F	2.1%	1.6%
other light	5.5%	4.3%
Total light	81%	85%
UO3	6.7%	5.3%
Bi2O3	5.0%	3.9%
ZrO2	3.3%	2.6%
Ce2O3	2.0%	1.6%
other heavy	2.2%	1.7%
Total heavy	19%	15%

The transformation of sodium salts of nitrate, nitrite, and carbonate to sodium hydroxide is straightforward. Following or in conjunction with these steps the wastes are calcined leaving a sodium hydroxide melt as a convenient medium for dissolving and mobilizing the remaining constituents of waste sludge. Such a scheme is illustrated in Fig. 3.

Caustic fusions are used extensively in analytical assay procedures for sample preparation prior to analysis. In fact, such fusions are typically used in Hanford's own assays of tank wastes. Thus, most species present in waste tanks are more or less soluble in such a medium. Previous work has shown that actual Hanford sludges can be readily prepared with such melts [Delegard, 1995].

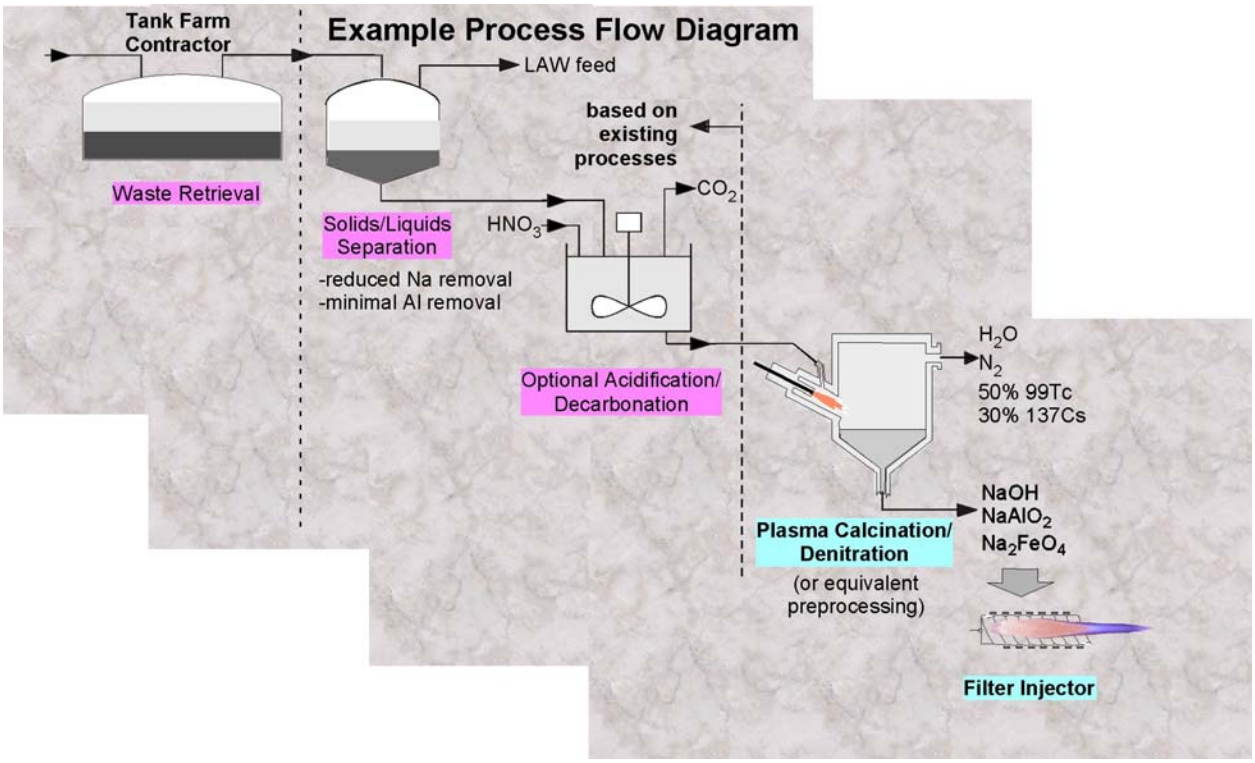

Fig. 3. Process flow diagram showing a pretreatment scenario that creates sodium hydroxide melt.

The flowsheet diagram illustrates the various stages anticipated for Archimedes feed preparation. Waste retrieval of sludges from tanks results in a 20 wt% slurry of waste oxides. This slurry undergoes sedimentation and decantation of the supernatant layer. The remaining sludge layer is further processed to denitrate and decarbonate the slurry before it is fed into a plasma calciner (or some other suitable calcination step) that produces a sodium hyroxide melt. The hydroxide melt is fluid in the range 400-800 C and can carry the waste oxide mixture to the filter injectors.

Injection and Vaporization
At the filter injectors, the hydroxide mixture is rapidly heated with a 5 MHz inductively heated thermal plasma. This sodium hydroxide torch is specifically designed to vaporize oxide mixtures carried by molten NaOH. The NaOH melt is nebulized at the injector and rapidly heated by the ~5000 C plasma in the torch. These inductively-coupled plasma torches operate solely on nebulized NaOH vapor without any added gas providing an extremely effective vaporization method.

Ionization and Separation
This high temperature vapor stream from the injector is directed into the Filter plasma at the center of the device. There the vapor is completely ionized by the main plasma, which is heated by helicon excitation at 5 MHz by two antenna assemblies that are integral to the device.

The Demo Filter is shown in Fig. 3 and is very similar in scale to the commercial version. A large vacuum chamber (a) within a modest magnetic field (b) of 500-1500 gauss contains plasma that is continuously fueled by injectors (c) and is composed of an oxide mixture that will represent waste. With a selected electrical potential (300-700 V) across the plasma and corresponding magnetic field correctly "tuned", heavy ions quickly spin out of the plasma and deposit on the heavy collector (d).

Light ions remain within the plasma and end up on the light collectors (e) at either end of the Filter chamber. Plasma is heated by two sets of RF (radio frequency) antennas (f) in each half of the Filter while the elements of the electrode/light collector assembly (e) maintain plasma electric field and vacuum pumps at either end and middle remove volatiles from both ends of Filter (g).

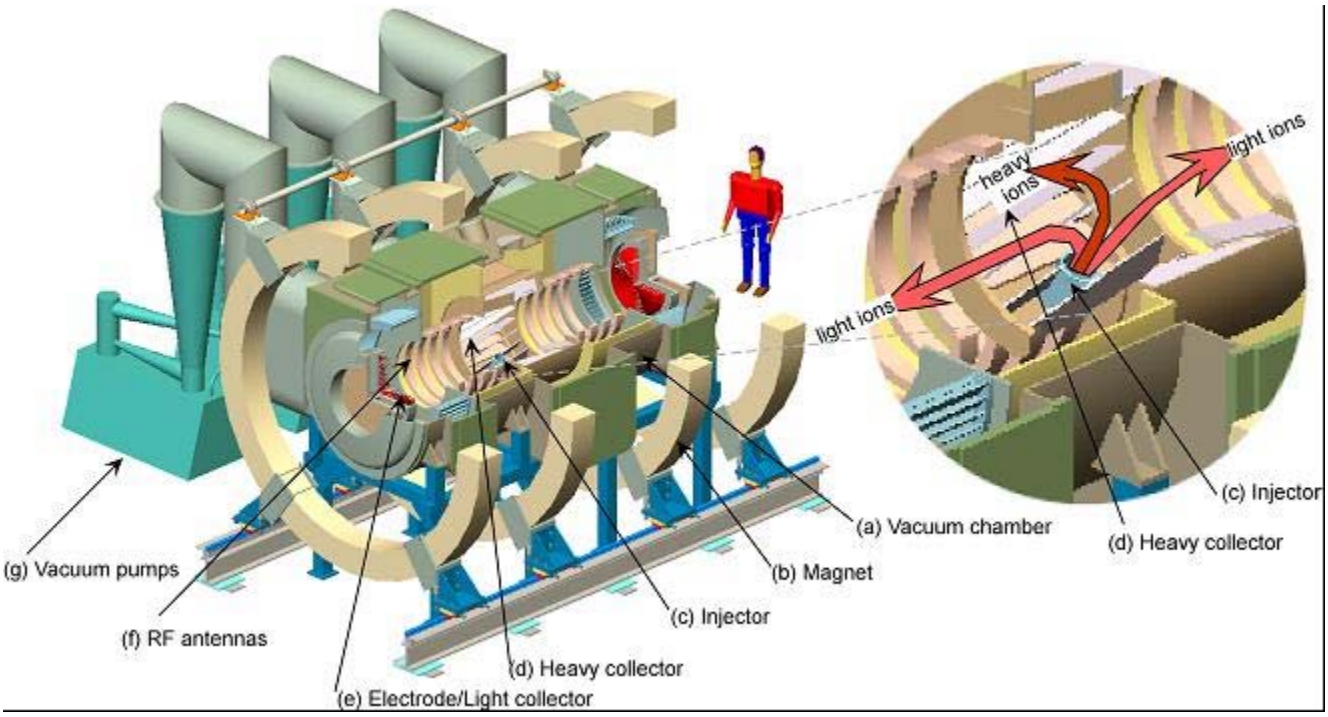

Fig. 4. Demo Filter schematic.

Heavy and Light Collection
Ions that exceed the tuned cutoff mass quickly spin out of the plasma and deposit in the heavy collector. Ions less than the cutoff mass remain in the plasma and deposit at the light collector/electrode assembly. Current design expectations suggest a throughput of around 0.7 MT/day of mixed oxides per Filter unit. This would produce 0.175 MT/day of heavy oxide and 0.525 MT/day of light oxyhydroxide. Oxygen and hydrogen atoms are more difficult to ionize that most other species and therefore will persist as neutral species. As a result, both heavy and light deposits will be fully oxidized and suitable for feed to either IHLW or ILAW processing.

One area of uncertainty in the Filter atomization of NaOH is the nature of "reconstitution" of NaOH on the Filter collector surface upon plasma condensation. Reconstitution of NaOH at the collector will need some migration and mixing of material at the collector surface. However, excessive mobility of atoms at the collector will lead to recycling of material back into the plasma. It is clear that optimal collection of NaOH will necessarily be a tradeoff between high enough mobility for reconstitution and low enough mobility to prevent excessive recycling of material back into the plasma.

Material input to the Filter will have a range of volatilities and therefore will require a range of temperatures for optimal Filter injection. Likewise, there will be a range of temperatures for collection of the vaporized material. Injection temperature is more or less defined by the most refractory material, which is likely to be ZrO_2, while collection temperature is defined by the most volatile species, NaOH.

The strategies for injection and collection are complimentary. For injection, rapid heating to very high temperature is essential to minimize fractionation or distillation of the materials being vaporized. This rapid heating occurs with a residence time adequate for even the most refractory materials to completely atomize. For waste collection, correspondingly rapid cooling is likewise needed to help minimize fractionation of plasma components. Collection, though, also needs time for reconstitution in order to minimize the production of small molecule volatiles such as O_2, H_2, and H_2O.

Reconstitution occurs in a domain at or near the collector surface where mobility is large enough to support surface atom transport but mobility is small enough to minimize atom recycling back into the plasma. Reconstitution should

be consistent with a collision rate large enough to support atom recombination into more stable molecules. This reconstitution period will in general be on the same order as the residence time for vaporization.

The material that accumulates at the light collector will be removed from the chamber as a sodium hydroxide melt while the heavy collector oxide will require a once per day cleaning operation. We will investigate a variety of options for that cleaning and plan to deliver the heavy oxide product as a water slurry to the HLW melter.

Modeling and simulation

The Filter is based on accepted principles of plasma physics and is therefore amenable to plasma modeling. In particular, we have developed an ionization model coupled with a Monte Carlo plasma simulation for selected ions in a background plasma. Ions whose mass exceeds the threshold are not confined by the plasma and escape to the heavy collector. Correspondingly, ions with mass less than the cutoff remain confined in the plasma and deposit at the light collectors at either end of the device. Because the Filter is a rather coarse ion separator, ions that fall within the filter edge end up in both the heavy and light collectors.

The ionization model allows us to use experimental ionization cross-sections and potentials to model the charge state of each species. Once ionized, the trajectory of each ion can likewise be modeled in the magnetic and electric fields of the Filter plasma and this forms the basis of the Monte Carlo simulation.

Furthermore, the ion and neutral radiation and conduction losses determine the power needed to sustain the plasma throughput. We have therefore also modeled those losses and used those loss estimates to predict the Filter power.

DEMONSTRATION PROJECT

Archimedes Technology Group, Inc. is engaged in a project to design, build, and test its ion Filter in a San Diego facility. This project will demonstrate the viability of the Filter to address Hanford's complex waste disposal problem. The demonstration will show:

1. Efficient separation of surrogate non-radioactive mixtures, initially sodium and bismuth oxides and then more complex mixtures;

2. Filter throughput of 0.13 mol/s feed rate for 0.8 m plasma diameter tandem device;

3. Energy cost per ion for NaOH plasma less than 500 eV per ion;

4. Filter tolerance for oxide complexity shown by following:

- inject oxy-hydroxide material, i.e. oxygen and hydrogen in feed;
- light element Na (sodium);
- heavy element Bi (bismuth);
- cutoff element Fe (iron);
- refractory element Al or Zr (aluminum or zirconium).

The demonstration facility building modifications have been completed and the Filter device is now being assembled. Current schedule calls for first results in early 2003.

Commercial Filter Nominal Specifications

The Filter shown in Fig. 3 is now being assembled as part of the Filter demonstration. The Filter for commercial application at Hanford and will be similar in scale to the Demonstration unit but will have the specifications as shown in Table III.

Table III. Commercial Filter specifications.

parameter	**value**	**comment**
Te	1.5 eV	electron temperature
chamber vacuum	100 mTorr	
rf power	8.0 MW per unit	demo is 4.0 MW
total power	14.0 MW per unit	demo is 7.0 MW
footprint	12' x 12' x 20'	hxdxl, per unit without vacuum pumps or power supplies, demo is slightly larger
processing rate	1.1 MT/day	for 50 wt% NaOH mixture with nominal Na/Al/Fe oxides as majority of light fraction
HLW oxide mass reduction	4.0	based on TWRS'99 SOUP projections for for BOM HLW feed

SUMMARY

We have described the novel Archimedes Filter and its application to high-level waste at Hanford and shown various model results showing Filter separation of ionized material. Such modeling provides an estimate the potential advantages of Filter tunability, both in cutoff mass (electric and magnetic fields) and in degree of ionization (plasma power). In addition, we have found that a Hanford waste oxide can be prepared as a sodium hydroxide melt and this melt can be vaporized by suitable ICP injectors in the plasma filter.

Oxide material at the heavy collector along with heavy radionuclides represents only a fraction of the total material input. This heavy oxide mixture is then ready to be vitrified in the HLW melter while the light oxide fraction can be disposed as LAW.

Developing a new technology is always challenging and many challenges remain in the Filter development, not the least of which is the eventual demonstration and commercial implementation of the Filter on radioactive waste at Hanford. We have confidence that our approach is a viable one and are anticipating that the Filter will become a part of the solution for Hanford's HLW disposal problem.

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