In Situ Cleanable HEPA Filter for Clean-Up of Vent Gas from HLW Waste Tanks

Bruce Bishop (bishop@ceramem.com; 781-899-4495)
Robert Goldsmith (goldsmith@ceramem.com; 781-899-4495)
Karsten Nielsen (nielsen@ceramem.com; 781 899-4495)
CeraMem Corporation
12 Clematis Avenue
Waltham, MA 02453

Duane Adamson (duane.adamson@SRS.gov; 803-725-5307)
Westinghouse SRS
Building 786-SA
Aiken, SC 29808

Presented at

Industry Partnerships for Environmental Science and Technology Conference
Morgantown, WV
October 30 – November 1, 2001

ABSTRACT

DOE has about 300 high-level waste liquid storage tanks at its various sites. These are maintained at a slight negative pressure to prevent leakage of radioactive contamination and avoid buildup of hydrogen in the headspace. The vent gas from these tanks is filtered through disposable glass-fiber HEPA filters. Replacement of these filters generates both a waste disposal problem and a problem associated with operator exposure during filter replacement.

CeraMem Corporation has developed a ceramic monolith filter with a composite structure as a long-lived replacement for disposal HEPA filters. The filter employs a fine-pored ceramic membrane barrier, which has HEPA filtration properties. Prototype full-size filters (18.5 ft^2 each) have been developed and supplied to Oak Ridge National Laboratory's HEPA Filter Test Facility for qualification testing and to the Savannah River Technology Center (SRTC) for simulated vent gas tests. Results presented in this paper show >99.97% retention in the standard 0.3 µm DOP aerosol challenge test (at ORNL) and the ability to be cleaned effectively by backflushing after plugging (at SRTC).

Continuing work in this program involves the possible supply of a full size vent gas filtration system for evaluation on a high-level waste tank vent line at the Savannah River Site (SRS). A decision on this demonstration installation will be made by DOE in FY 2002.

LIST OF ACRONYMS

ATI	Air Techniques International
DOE	U.S. Department of Energy
DOP	di-octyl phthalate
FTF	Filter Test Facility
HEPA	high efficiency particulate air
HFTA	HEPA filter test apparatus
HLW	high-level waste
INEEL	Idaho National Engineering and Environmental Laboratory
ORNL	Oak Ridge National Laboratory
SC	South Carolina
SRS	Savannah River Site
SRTC	Savannah River Technology Center

I. BACKGROUND

Conventional disposable glass-fiber HEPA filter cartridges are used throughout the U.S. Department of Energy (DOE) complex in various radioactive process systems. These filters require routine removal, replacement, and disposal. This process is not only expensive, but subjects site personnel to radiation exposure and adds to an ever-growing waste disposal problem. The conventional HEPA filters also have safety concerns regarding the strength of filter media, water damage, and operation in environments with elevated temperatures. The Defense Nuclear Facility Safety Board issued a report entitled "HEPA Filters used in the Department of Energy Hazardous Facilities", DNFSB/TECH-23, in which these and other concerns pertaining to conventional HEPA filters were addressed. (1)

The SRS high-level waste (HLW) tanks are designed to hold approximately 1.0 million gallons of radioactive liquid waste. Figure 1 depicts a typical HLW tank ventilation flow diagram. The tanks are located outdoors and buried in the ground except for the tank top. They are equipped with a ventilation system that provides approximately 500 cfm of filtered air within the headspace. The airflow maintains the tank contents at negative pressure (-1.0" water column). The air flows through a demister and condenser where water is removed from the air stream. The flow then enters a reheater where the air is heated above the dew point before being filtered at the outlet HEPA Filter and released to the atmosphere. Tanks with secondary containment also have a separate ventilation system for the annulus space. The negative pressure maintained on the tank contents prevents the release of radioactive material to the environment. However, it also allows for possible atmospheric dust to be pulled into the tank during normal operation.

The glass-fiber HEPA filter must exhibit a particle removal efficiency of 99.97+% when challenged by thermally generated di-octyl phthalate (DOP) aerosol with a diameter of approximately 0.3 microns. The pleated glass-fiber HEPA filter media has approximately 240 ft^2 of surface area and is typically contained in a 2 ft x 2 ft x 1 ft housing and exhibits a 1 inch of water column (w.c.) differential pressure (ΔP) across the filter media when clean. A HEPA filter remains in service until the filter media reaches a predetermined maximum pressure drop (approximately 5 inches w.c.) or a high source term due to radioactive buildup, and then the filter is replaced.

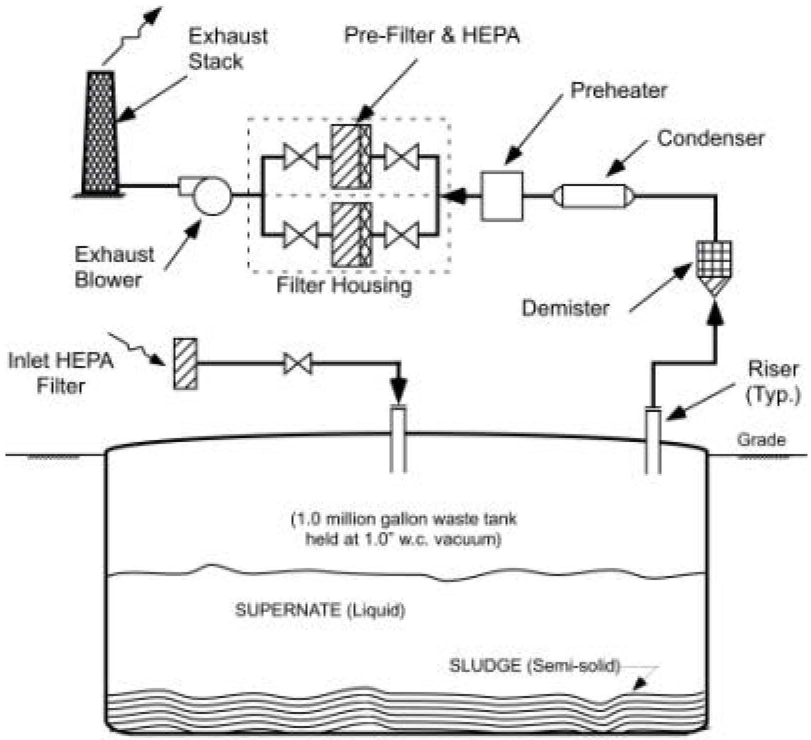


Figure 1. Typical HLW Ventilation System Flow Diagram

SRS requires that all process HEPA filters pass an in-place leak test both before being placed into service, and periodically thereafter. DOP test connectors are designed into each HEPA installation to facilitate the routine in-place DOP test per national and site standards. Both the material challenge and the filter efficiency tests conducted on the filters in this study were designed to simulate conditions found in HLW tanks at SRS.

Previous research has been conducted at other DOE sites such as Lawrence Livermore National Laboratory and Oak Ridge National Laboratory to develop an *in situ* cleanable HEPA filter with high media strength. W. Bergman, et al. conducted research on various filter media, such as steel fibers, ceramic, and sintered metal, using reverse air pulse as the *in situ* cleaning method.(2) The results of these investigations indicate that commercially available filter media could be applied to the development of an *in situ* cleanable (using reverse air pulse) HEPA filter system that would meet the performance criteria established for a conventional HEPA filter system.

In FY98 SRS started working on the theory of using high-pressure drop filter media in an *in situ* regenerative filtration system. The *in situ* cleaning of the filter media used an aqueous solution to regenerate the media to a new, clean filter status. Testing conducted in FY98 and FY99 indicated that an *in situ* cleanable filtration system is feasible for use on the HLW tanks.

A cost analysis conducted on cleanable HEPA filters showed that the *in situ* cleanable filters have the potential of being cost effective when compared to the conventional glass-fiber HEPA filters. The study estimated that the DOE complex uses an average of 11,000 filters per year at an estimated cost of $55 million. Using *in situ* cleanable HEPA filters could save the complex $42 million a year in operating cost. The study did not include costs associated with glass-fiber filter breakthrough or rupture, but these additional costs increase the cost benefit of the *in situ* cleanable filters for the complex.(2)

The DOE Tank Focus Area and the National Energy Technology Laboratory issued a "Call for Proposals" to identify vendors conducting research in the area of *in situ* cleanable/regenerative filters or vendors interested in pursuing such technology.(3) A technical evaluation was conducted on the proposals. Based on the proposals reviewed, CeraMem Corporation, located in Waltham, Massachusetts was selected to support this research program. This project is now in its third year. In the initial phase of the program, CeraMem developed small test units (lab scale filters) that demonstrated HEPA filter performance and ability to be regenerated by backflushing. The results of this phase were reported at last year's conference.(4) This paper presents results from this past year's activities, which entailed the development and testing of full-size, prototype filters.

II. TECHNOLOGY

Figure 2 depicts the design of the CeraMem filter media and the air flow passage through the "dead-end" monolith filter. The passageway ends are plugged with ceramic cement in an "alternate, checkerboard pattern". As shown, the dirty air with particulate matter enters the filter media and the gas flow is constrained to pass through the monolith walls separating inlet and outlet passageways. The membrane coating covers the passageway walls, and the entrained particles are filtered from the air onto the surface of the microporous membrane. The clean air flows down the outlet passageways and is exhausted from the filter. Figure 3 shows a full-size prototype filter.

Below are the characteristics of the "full-size" ceramic monolith filters tested in this phase of the program:

• Monolith Membrane Support:	Silicon carbide
• Membrane Coating:	Glass-frit-bonded zirconium silicate
• Overall Element Dimensions:	5.66" diameter x 12" long (144 mm x 305 mm)
• Monolith Cell Size:	0.079" (2 mm)
• Monolith Cell Wall Thickness:	0.0315" (0.8 mm)
• Filtration Surface Area:	18.5 ft^2 (1.72 m^2)

Figure 4 shows photomicrographs of the silicon carbide (SiC) monolith pore structure (left) with a membrane surface coating (right), with a pore size between 0.2 – 0.5 μm. This ceramic membrane provides a relatively low-pressure drop while obtaining HEPA filter retention efficiency. Figure 5 is a photomicrograph of a cross-section view of the SiC monolith with the membrane coating.

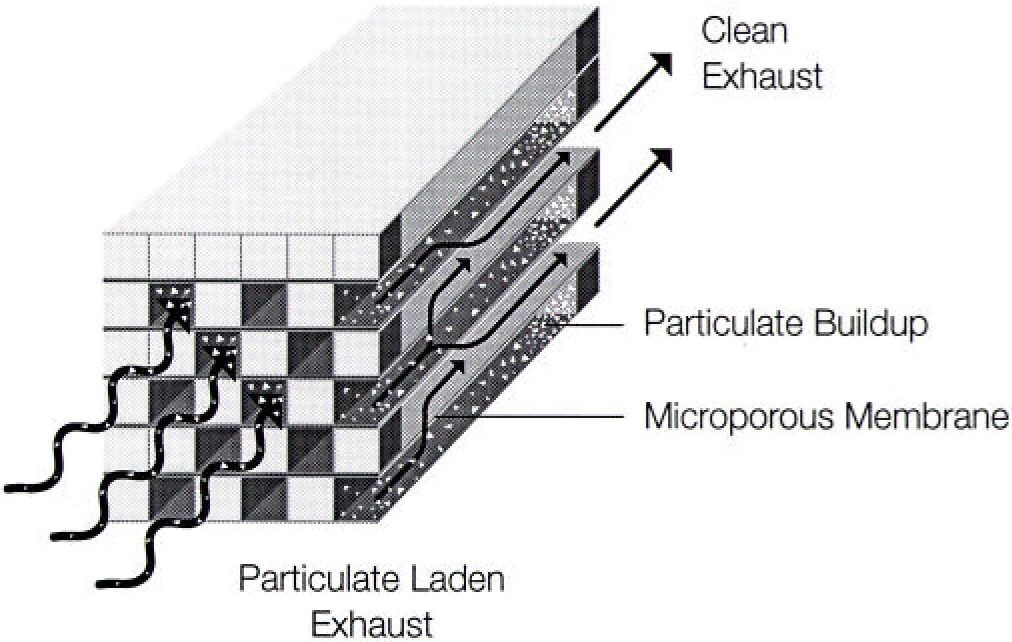


Figure 2. Airflow Pattern Through Ceramic Monolith Filter

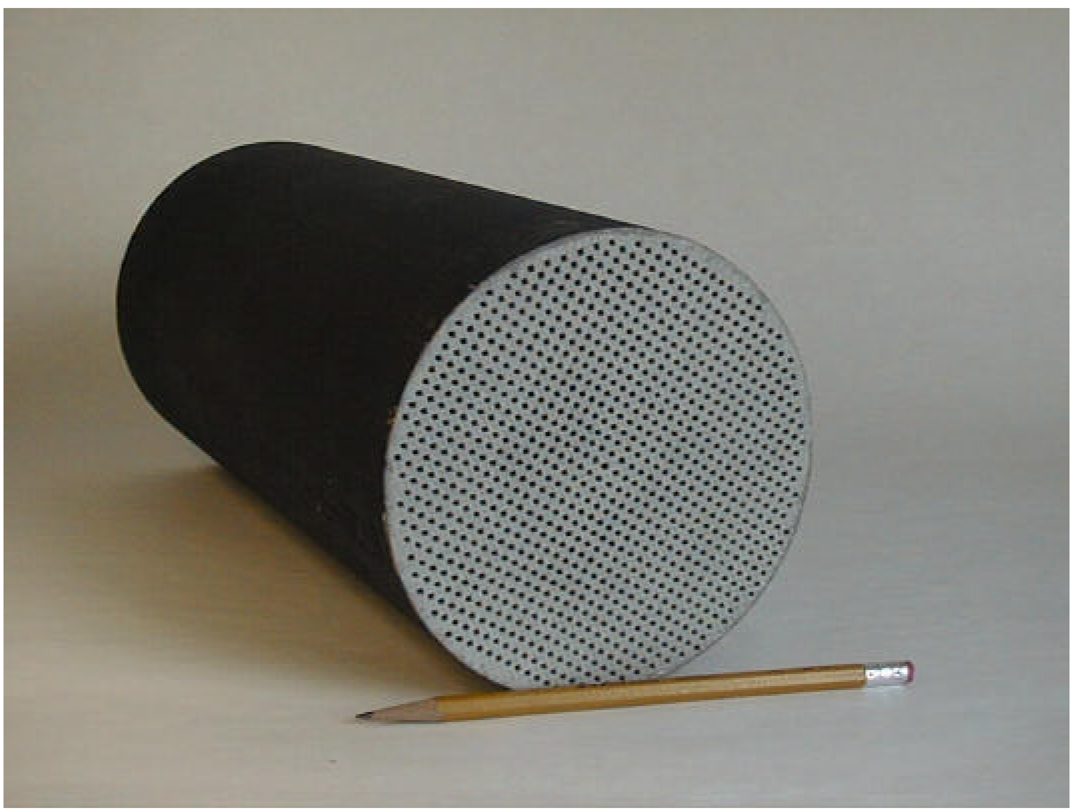


Figure 3. Photograph of Full-Size Monolith Filter (5.66” diameter x 12” length)

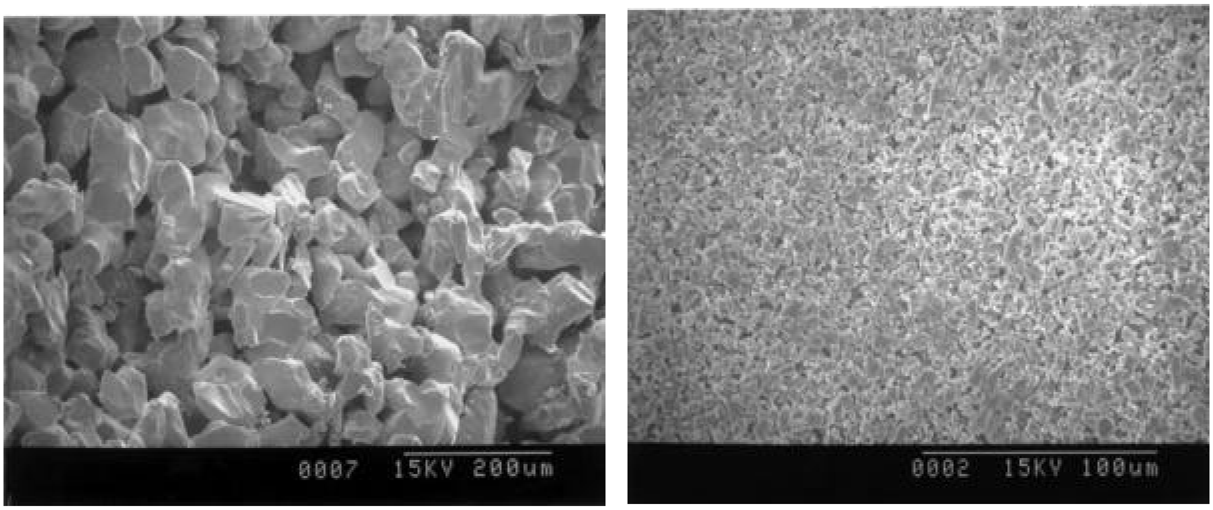


Figure 4. Surface View of Monolith Wall Pore Structure (left), and Overlying Membrane Surface Pore Structure (right)

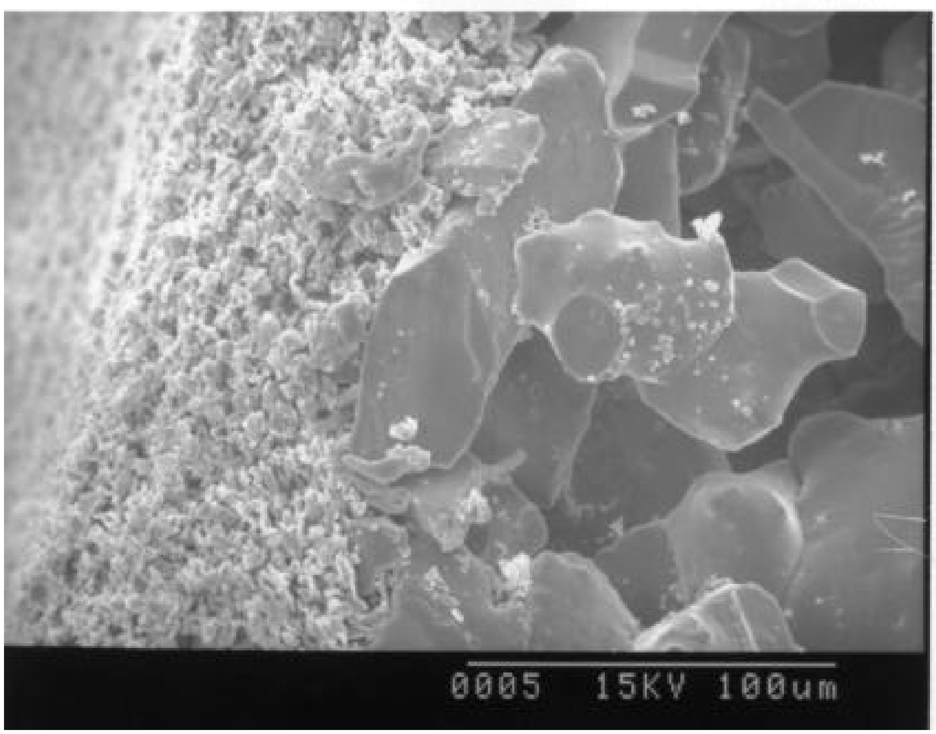


Figure 5. Cross-Sectional View of Membrane/Monolith Wall Structure

III. FULL SIZE FILTER HEPA CHALLENGE TESTS AT ORNL

Two series of filters were shipped to ATI to conduct filter efficiency tests at the Oak Ridge FTF. ATI modified their test equipment to conduct these efficiency tests in early 2001. Filters were supplied over the spring and summer of 2001 for DOP challenge tests. The second series of thirteen filters, with the preferred membrane coating, were tested under two different gas flow conditions and the results are given in Tables I and II.

Table I. DOP Retention Data for Full Size Filters

Filter Serial No.	Air Flow, cfm	% Retention (Pass)	% Retention (Fail)
H-28	25	99.999	
	125	99.982	
H-29	25		98.5
	125		95.9
H-30	25		99.67
	125		93.2
H-31	25	99.999	
	125	99.986	
H-33	25		99.28
	125		96.4
H-39	25		97.6
	125		94.2
H-40	25		99.44
	125		96.8
H-41	25	99.999	
	125	99.993	
H-42	25	99.999	
	125	99.984	
H-43	25	99.999	
	125	99.989	
H-44	25	99.999	
	125	99.986	
H-46	25		99.05
	125		97.9
H-47	25	99.999	
	125	99.992	

Table II. Filter Pressure Drop at Design Flow (125 cfm)

Filter Serial No.	Pressure Drop, inches water
H-28	26.1
H-29	26.0
H-30	21.2
H-31	22.5
H-33	27.2
H-39	26.8
H-40	26.5
H-41	25.2
H-42	25.6
H-43	25.1
H-44	23.2
H-46	26.8
H-47	22.6

As can be observed in the data of Table I, seven of the thirteen filters tested passed the HEPA challenge test readily. Six failed, with quite consistent retention levels. As of the date of preparation of this paper, the filters have not yet been returned to CeraMem for examination of what appears to be a recurring defect. In any case, at this stage of making the prototype filters, the yield in excess of 50% is considered to be acceptably good.

Furthermore, the pressure drop data in Table II are acceptable for a full flow system. It may be noted, however, that these pressure drop levels are substantially higher than is the pressure drop of standard, disposable, glass-fiber HEPA filter elements.

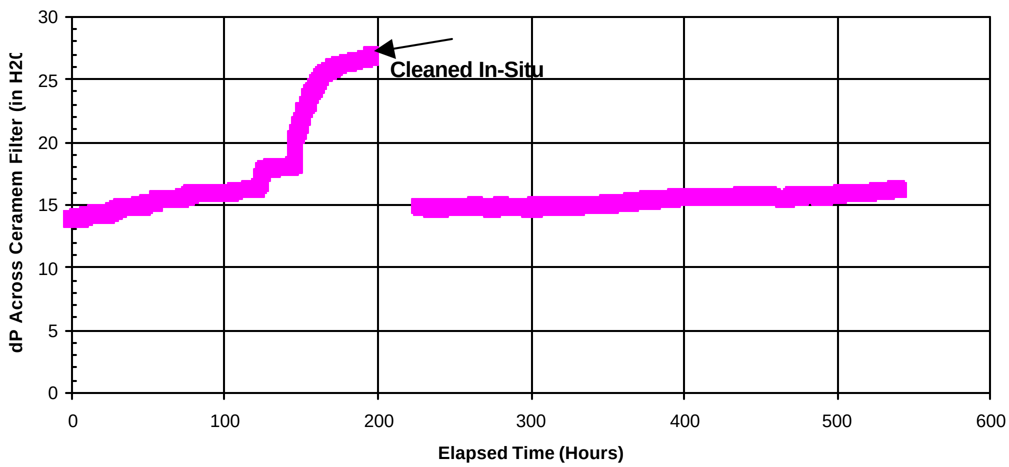
IV. PROCESS TESTS AT SRTC

Several filters have been supplied to SRTC. Initial tests with simulated waste sludge follow the test procedures described in the paper presented at last year's conference with lab-scale filter elements and are not described again here.(4) In initial tests with one filter, using moderate particulate loadings, the filter has been repeatedly plugged and backwashed successfully to recover the initial pressure drop performance.

A CeraMem filter that was subjected to such preliminary simulant testing at SRTC was re-tested by ATI at the FTF for particle retention. The results are positive in that there was little or no change in the retention capability after many *in situ* cleaning cycles. This indicates that the filter media is not deteriorating when undergoing repeated cleaning.

Data showing one filtration cycle (with accelerated plugging during the initial filtration cycle), followed by cleaning and a subsequent filtration cycle are shown in Figure 6.

Figure 6. Full Size Filter HLW Sludge Challenge Test



V. <u>RELATED APPLICATION AT INEEL</u>

A set of similar, but smaller diameter, filters has been supplied to INEEL for tests. Three filters have been installed in a filter vessel and are being tested with a simulated non-radioactive calcine material to obtain results for cleaning the filters *in situ* using a reverse air pulse cleaning method. The material is very dry throughout the transfer (as well as the air used to backpulse). The clean filter pressure drop of 10" w.c. increased to approximately 18" w.c. before online backpulse regeneration. The filters regenerated/recovered very well *in situ* from the air backpulse method. After twenty (20) plugging/cleaning cycles, the filters regenerated to a clean filter status.

VI. <u>ALTERNATIVE FILTRATION FULL SCALE SYSTEM DEPLOYMENT</u>

A full-scale Alternative Filtration System is planned for construction with the hot (i.e., radioactive) demonstration of the system being conducted at SRS. Before the hot deployment, the full-scale system will undergo cold testing at SRTC.

Figure 7 shows the filter housing containing the seven ceramic filters required to handle the total vent gas flow, of up to 800+ acfm. The filter vessel will be incorporated into the skid design previously developed by SRS, replacing the standard HEPA filter. The skid blower size will be increased to handle the increased pressure drop of the ceramic filters. The dirty air will be drawn into the bottom end of the housing and clean air will exit through the top of housing. After the filter becomes plugged with particulate or when the radioactivity from the accumulated particulate approaches area limits, the *in situ* cleaning system will be activated. The elements will be cleaned *in situ* via back flushing an aqueous cleaning solution through the media, with the backflush liquid returned to the HLW tank.

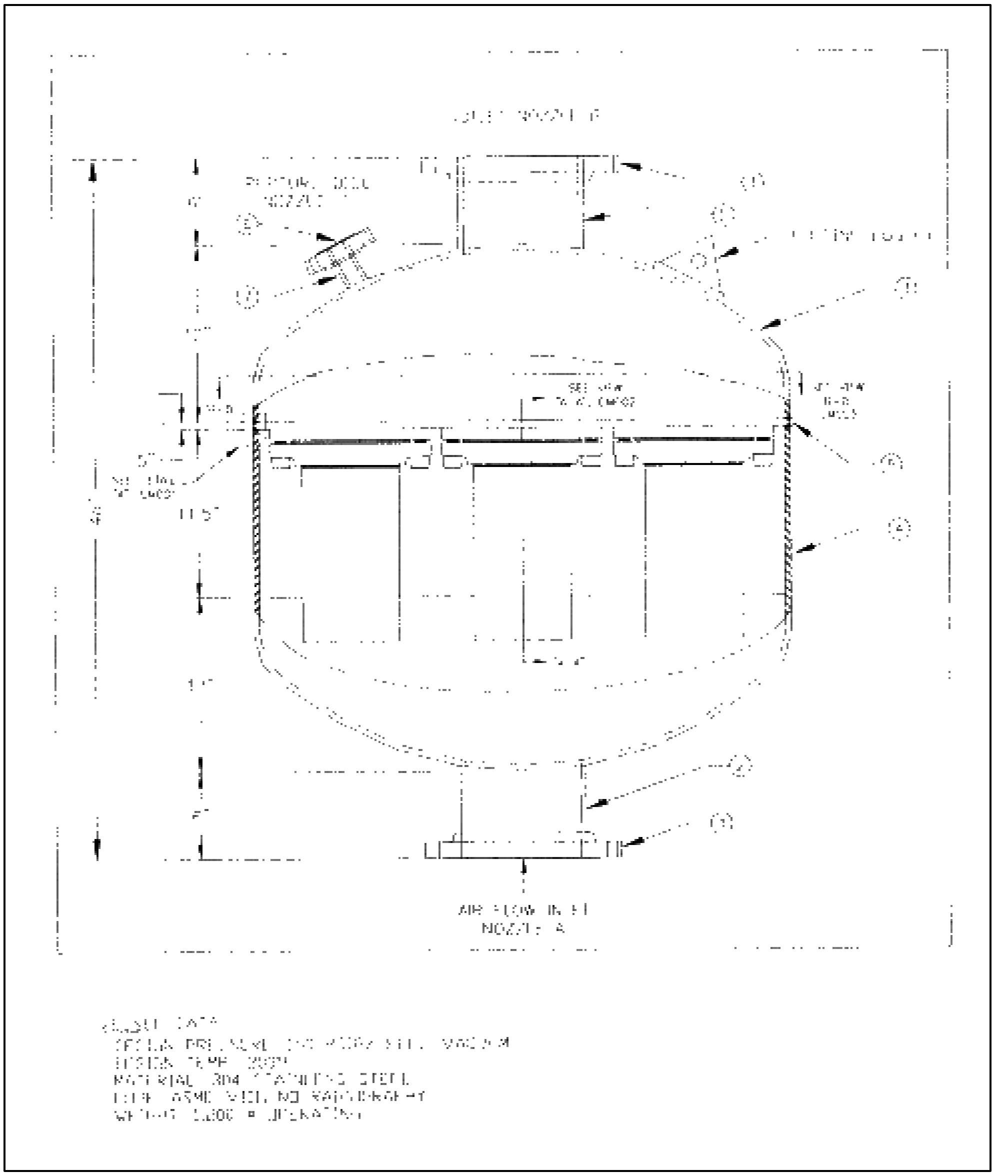


Figure 7. Filter Housing Containing Seven Filter Elements (800+ acfm)

VIII. CONCLUSIONS

Alternative media such as CeraMem ceramic membrane filters holds great promise for use as an *in situ* cleanable/regenerative HEPA filter. The proof-of-principal testing has shown that the ceramic media could be suitable as an alternative HEPA filter media. During simulated testing the filter regenerated well *in situ* with a potential for 15 years life under actual field conditions of the HLW tanks. In addition to eliminating the costs associated with disposing of and replacing disposable filters, the strong filter media also reduce the potential of a catastrophic HEPA filter failure due to rupture of the filter media.

The CeraMem ceramic membrane filter technology has been scaled up to produce full size filters. These have been shown to pass the HEPA retention test requirement in independent tests at ORNL's FTF. Process tests at SRTC show that the filter can be plugged with simulated particulate matter and regenerated by backflush cleaning. Initial test results show minimal (if any) loss of retention with repeated plugging and cleaning.

REFERENCES

1. Defense Nuclear Facilities Safety Board Technical Report, DNFSB/TECH-23, May 1999.

2. Bergman, W., Larsen, G., Lopez, R., Wilson, K., Witherell, C. and McGregor, M., "Further Development Of The Cleanable Steel HEPA Filter, Cost/Benefit Analysis, and Comparison With Competing Technologies" 24th DOE/NRC Nuclear Air Cleaning And Treatment Conference, July 15- 18 1996

3. Technical Task Plan, TFA Tank Waste Retrieval and Closure, "Alternatives Filtration Technologies" TTP #: SR1-8-WT-21 9/23/99

4. Goldsmith, R., Bishop, B, Nielsen, N, and Adamson, D., "*In Situ* Cleanable HEPA Filter for HLW Waste Tanks", Industry Partnerships for Environmental Science and Technology Conference, Morgantown, WV, October 17, 2000