Development of a Catalyzed Ceramic Filter for Combined PM2.5 Removal and VOC and CO Oxidation

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1. PROJECT BACKGROUND

The Department of Defense has a large number of particulate emissions sources that may be impacted by new PM2.5 regulations. The USEPA has proposed new PM2.5 regulations in addition to those already enacted to control PM10 because it believes that current standards do not sufficiently protect the public health. The proposed new standard presents a clear challenge to many industrial and Governmental segments of our society. Indeed, it is questionable whether technology exists to control PM2.5 economically from the majority of sources.

The Defense Department, through the Strategic Environmental Research and Development Program, has identified emissions sources that need enhanced control systems if proposed regulations become reality. These sources include engine test cells, diesel power generation sets (gensets), shipboard diesel engines, heavy-duty mobile diesel sources, trash incinerators, medical waste incinerators, demil waste incinerators, and steam boilers. These sources are emitters of fine particulate matter, potentially subject to the new PM2.5 standard, and many are emitters of significant amounts of volatile organic compounds (VOCs) and carbon monoxide (CO).

2. PROJECT OBJECTIVES AND PROJECT OVERVIEW

The focus of this project is the development of a novel technology for PM2.5 removal with substantially 100% removal efficiency. The technology is a highly compact, ceramic-membrane-coated monolith filter. Moreover, this same filter can be coated with non-selective oxidation catalysts for the combined, simultaneous removal of particulate and vapor-phase organics and carbon monoxide. While no single emission control technology will address all of DoD's needs in this area, this filtration technology may be able to address specific needs. In order to address these needs, the following technical objectives were originally established for this project.

a) The filter is to be based on the use of porous, honeycomb silicon carbide monoliths as macroporous supports (15-25 micron mean pore size) for thin ceramic membrane coatings with a pore size of about 0.1 to 0.2 μm. This composite filter is to have a low-pressure drop, ca. 15 inches water at a face velocity (equivalent to "air to cloth ratio") of 10 ft/min. A virgin membrane-coated filter is to have greater than 90% retention by weight of gas-entrained particles between 10 micron and 30 nm. A conditioned filter is to have greater than 95% retention by weight of gas-entrained particles in the same size distribution.

b) One possible modification of the composite filter is to have an additional overlying layer of a low-temperature oxidation catalyst suitable for continuous "passive" soot oxidation in engine exhaust streams, active at exhaust gas temperatures below 400°C. Oxidation of soot would allow for passive recovery and/or maintenance of the filter pressure drop during operation. This catalytic filter would be used for PM2.5 removal from engine combustion sources in which the particulates are primarily organic in composition. Catalyst addition is not to increase total pressure drop over the target given above.

c) A second possible modification of the composite filter is to have a low-temperature oxidation catalyst, impregnated inside the pores of the filter, suitable for continuous oxidation of hydrocarbon vapors and CO at exhaust gas temperatures below 300°C. The catalyzed filter should have more than 90% destruction of a model VOC and CO at a gas hourly space velocity of 40,000 hr^{-1} or higher. This catalytic filter would be used for

PM2.5 removal from combustion sources in which the particulates are primarily inorganic in composition. Catalyst addition is not to increase total pressure drop over the above target.

Over the course of this past year, these product development objectives have been modified as more has been learned about specific DoD needs. These are given below, as a restatement of the above objectives.

a) Depending on the specific emission source, filters with different retentions will be desired. These can range from HEPA filters (99.97% removal of 0.3 μm DOP aerosol), with a relatively high pressure drop acceptable, to diesel exhaust traps (>95% soot removal for virgin filter), but with a relatively low pressure drop. Thus, the development of a family of membrane coatings with a trade-off between retention and pressure drop has become a project objective.

b) The application of a soot oxidation catalyst overlying the ceramic membrane coating as a desirable product variant for a diesel trap no longer appears feasible. This is due to the high sulfur content in DoD diesel fuels, which is foreseen to remain unchanged even as the non-military fuel supply moves to a low-sulfur fuel. The high sulfur content of DoD diesel fuel is problematic in that sulfate make and catalyst poisoning by the catalyst used in catalyzed traps (precious metals) precludes this product technical approach. Accordingly, the development of a diesel trap, based on the high-efficiency membrane-coated filter is now focused on two regeneration approaches: use of metal oxide fuel borne catalysts (FBC's) or thermal, oxidative regeneration.

c) The third product variant above, deposition of an oxidation catalyst within the pores of the filter, now appears less desirable than the use of a monolith channel-flow oxidation catalyst downstream of the filter. This is based on the lead application originally foreseen for the catalyzed filter, i.e., incinerator off gas. A preferred approach for incinerator off gas treatment is filtration at an elevated temperature (>500°C) to remove ash that catalyzes dioxin formation, and to use a downstream oxidation catalyst at a lower temperature for organics/CO oxidation. The lower temperature is required for catalyst stability.

Accordingly, the product development activity now is directed toward the development of:

a) A range of membrane coated filters with a family of membranes providing a tradeoff between membrane retention and filter pressure drop, and

b) Processes in which such filters are combined with soot or gaseous contaminant oxidation catalysts in manners other than by deposition of a catalyst on or within the filter.

3. SUMMARY OF TECHNICAL APPROACH FOR MEMBRANE-COATED MONOLITH FILTERS

CeraMem has previously developed and commercialized membrane-coated, monolith-based gas particulate filters. The flow pattern and location of the membrane coating for these filters are shown in Figure 1. These earlier filters were based on cordierite honeycomb monoliths and had limited particle removal efficiency due to two reasons. First was the thermal expansion mismatch between the monolith and membrane, leading to membrane cracks. Second was the presence of large "pores" in the monolith wall, leading to pinhole membrane defects.

Figure 1. Dead-Ended Gas Particulate Filter Construction and Gas Flow
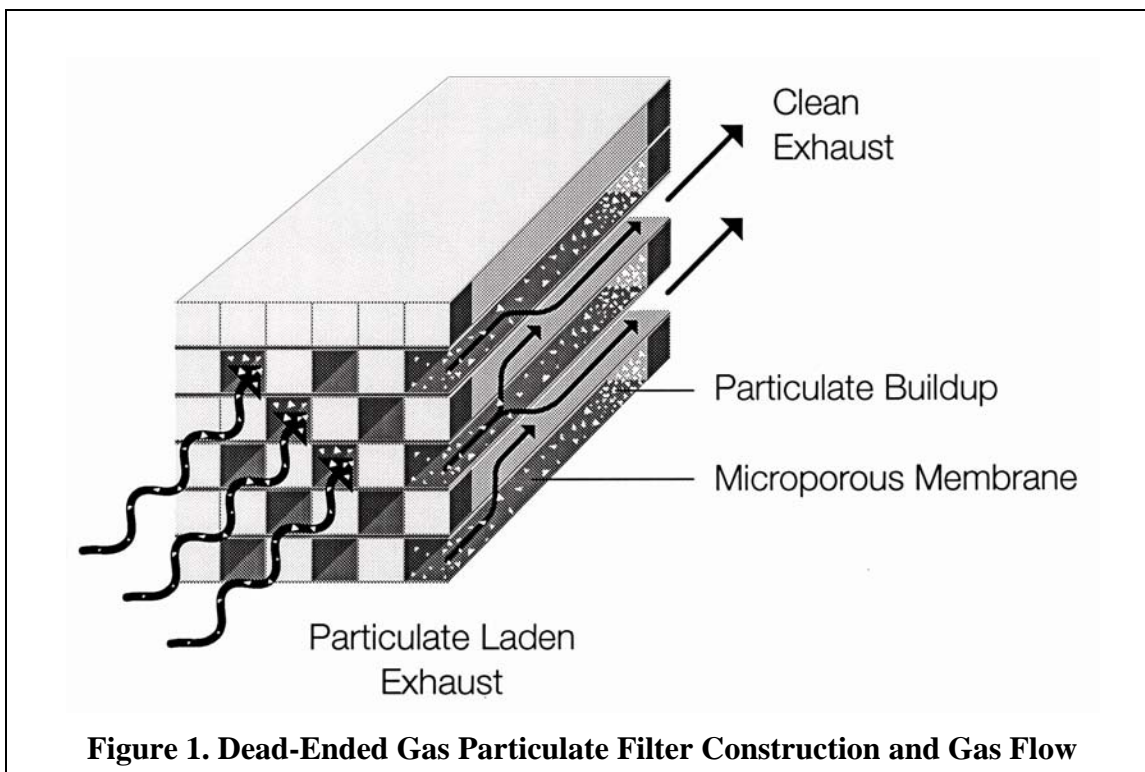
To overcome these limitations, a new filter, based on silicon carbide (SiC) monoliths, is being developed in this program. SiC has a larger thermal expansion coefficient (CTE) that better matches the CTE of membrane coatings, avoiding crazing cracks formed during membrane firing. In addition, the pore size distribution of the SiC is much sharper than that of cordierite, eliminating membrane pinhole defects. In addition, the very high permeability of the SiC monolith material can provide for a lower pressure drop filter.

4. PROJECT ACCOMPLISHMENTS IN 2000

Project results during 2000 are described in sections below according to task.

FY99: Task 4: Technical and Economic Feasibility Analysis

This task was completed during 2000. Three potential applications were analyzed for preliminary economic feasibility, and a report, submitted previously to SERDP, is included in Appendix A.

FY00: Task 6: Develop Full-Size, High-Efficiency Particulate Filters

During the first project year (1999) a membrane was developed that combined low pressure drop with high retention for particles in the size range 30 nm to 10 μm. This was accomplished by forming two membrane layers, each from silicon carbide (SiC) powders of narrow particle size distribution. On firing, the SiC powders bonded due to the formation of a surface silica layer on the SiC particles. Details of this research are included in the Annual Report for 1999.

During this second year of the project, it proved difficult to scale this membrane formation procedure to larger, full-size filters (diameter of 5.66", length of 12", and about 20 ft^2 filter area). This was in part due to difficulty in controlling the firing conditions so as to achieve a consistently, strongly bonded SiC membrane. This led to the development of two significant changes in membrane formulation, which led to substantially improved membranes.

- First, a glass frit was added to the SiC membrane slip formulation to melt and bond the membrane forming particles. Several frits were examined, at different proportions to the SiC powder and fired under different firing profiles. Membranes were formed which were reliably and strongly bonded. However, control of the membrane formation procedures proved difficult to control in terms of porosity and defects, related to reactivity of the SiC powder with the frits.

- Zirconium silicate powders (zircon) were substituted for the SiC powders, to eliminate the SiC reactivity problem, and a single layer membrane was developed (single firing cycle vs. two for SiC two-layer membranes).

The development of the new glass-bonded membranes has focused on maintaining high particulate removal efficiency and at the same time reducing the membrane pressure drop as much as possible. The ranges of filter performance sought have been between the following:

- HEPA filters, with a pressure drop of 20" H_2O at a filter face velocity of 10 ft/min (at room temperature)

- Diesel filters, pressure drop <10" H_2O at a filter face velocity of 10-15 ft/min (at room temperature), with high soot removal efficiency (>99%)

In order to develop a family of membranes between these retention/pressure drop ranges, it was necessary to continue work on smaller size filters before progressing to applying the membrane coatings to full size filters. The results below, therefore, are both for smaller filter coupons as well as full size filters.

Membrane Formation Formulations and Procedures

A significant matrix of different variables was explored in the first half of 2000. Rather than catalog these, the final membrane formulation and processing conditions selected will be presented. The procedures for slip casting membrane layers procedures were similar to that detailed in the previous annual report, except that the inorganic materials have been changed in accordance with the outline above. Firing of the membranes included a firing ramp at 5°C/min, a hold step for burn out of the pore formers (550°C), followed by heating to bond the membrane at 1150°C to 1190°C, with a hold at temperature for 10 minutes. Table 1 lists the constituents used in the membrane slips found suitable for coating the SiC monolith supports. Table 2 shows the composition ranges of the slips.

Filter Evaluation (at CeraMem)

Filter testing was performed on monoliths coated with ceramic membranes and plugged in the "checkerboard"-plugging plugging pattern of Figure 1. Measurements were made of (a) pressure drop, as a function of gas flow rate, and (b) particulate retention as a function of particle size.

The pressure drop was determined as a function of gas flow in a test setup in which a downstream blower drew ambient air through the filter. The flow rate was measured by a flowmeter located downstream of the filter. A static pressure tap was located between the filter and the flowmeter and attached to one leg of a U-tube manometer to measure filter differential pressure. A throttling valve between the flowmeter and the vacuum cleaner controlled gas flow.

Table 1. Casting Slip Components for Glass Bonded Zircon Membranes

Slip Component	Material
Liquid Vehicle	Ultrafiltered water
pH Modifier	NH$_4$OH (Aldrich Chemical Co., Milwaukee, WI)
Pore Former Dispersant	Lomar PWA (Henkel, Amber, PA)
Pore Former	Carbon Black Monarch 120 (Cabot Corp., Billerica, MA) Activated Carbon WPH pulverized (Calgon Carbon Corp. Pittsburgh, PA)
Membrane Inorganic	Zircon TAM-FM Zircon powder TAM-GZM Zircon powder (Cookson Matthey, Buffalo, NY)
Membrane Inorganic Binder	AL-Si-Glass Pemco glass frit, P-941-P (Trinity Ceramic Supply, Dallas, TX)
Antifoam Agent	Antifoam 65 (Dow-Corning, Midland, MI)
Viscosity Modifier	Polyethylene Glycol 20M (Union Carbide Corp., Danbury, CT)

Table 2. Typical membrane casting slip formulation

Slip Component	Mass	Concentration Basis
Ultrafiltered Water	94.0-100 g	Solids volume content = 0.279
NH$_4$OH	0.10 g	To pH ≈ 9.5
Lomar PWA (48% active)	0.32-1.57 g	5-11 wt% on carbon + dispersant
Carbon Pore Former	6.22-13.77 g	12-35 vol% on inorganics
Antifoam	0.09-0.1 g	0.1 wt% on water
Zircon, TAM-FM	23.3-28.0 g	40 vol% of Zircon
Zircon, TAM-GZM	35.0-41.9 g	60 vol% of Zircon
Pemco, P-941-P	19.44-23.26 g	37 vol% on inorganics
PEG 20M	6.1-6.5 g	Ca. 6 wt% on water

Particle retention was measured by filtering aqueous suspensions of a narrowly sized silica powder with 1.5 μm diameter. The test suspension had a turbidity of approximately 800 NTU (normal turbidity units). The suspension was pumped through a filter and the flow rate of the suspension was adjusted to give a constant filtration face velocity (0.03 ft/min) through the filters. Filtrate samples were collected initially and over 20-second intervals as filtrate was removed. The turbidity of filtrate aliquots was measured and retention calculated relative to feed suspension turbidity. For the results presented below, the filtrate turbidity was used to determine filter retention by the formula:

$$\% \text{ Retention} = 100 * (\text{Feed NTU} - \text{Filtrate NTU})/(\text{Feed NTU})$$

For the results presented, the retention associated with the *initial* filtrate sample is reported since the buildup of a filter cake during testing increased retention and differences in intrinsic membrane performance became less discernable.

HEPA Filter Tests at Other Sites

The CeraMem liquid filtration retention test was calibrated with a more meaningful gas filtration test. This gas filtration test is a standard "HEPA" filter test, in which a filter is challenged with a dilute fine aerosol and retention measured with optical instruments. Three test organizations were used to evaluate the filter elements. First was the Energy and Environmental Research at the University of North Dakota (EERC). The second was the Westinghouse Savannah River Site Technical Center (SRTC) in Aiken, SC. The third was AIR TECHNIQUES International (ATI) in Baltimore and their test facility at Oak Ridge National Laboratory, where ATI routinely tests HEPA filters for the DOE Weapons Complex prior to filter installation. EERC uses a polydisperse NaCl aerosol; SRTC uses a polydisperse poly (alpha-olefin) (PAO) aerosol; and ATI uses a monodisperse dioctyl pthalate (DOP) aerosol. Photographs of the ATI test system at ORNL used for full size filters are contained in Appendix B.

Results: Determination of Retention Limiting Defects

In the course of this year, an important observation was made relative to what controls the retention characteristics of the membrane-coated filters. Specifically, for most filters, defects at the ends of the monolith are the passage points for particles. Early measurements to relate pressure drop with retention led to poor correlations. Once the defect location was found, a technique to seal the ends of the filters was developed and implemented. It consists of the following steps:

- After applying the ceramic membrane and plugging the filter passageways at the ends to achieve the dead-end flow configuration, both end faces are dipped into diluted passageway cement. This coating forms a continuous layer over the end faces and a short distance into the filter passageways. The coating is fired at 500°+C at which temperature, the colloidal silica bonds the SiC particles. The recipe for the end-coating cement is given in Table 3.

Table 3. Composition of End-Coating Cement

Component	Wt. %
Water, pH=10	24.3
Antifoam, DC-65	0.01
Viscosity modifier, PEG 20M	14.5
Colloidal Silica, Ludox HS 40	12.2
Silicon Carbide, H.C. Starck UF10	6.9
Silicon Carbide, FEPA grit 1200	17.3
Silicon Carbide, FEPA grit 320	24.8
Total	100.01

- This is followed by dipping the end faces into a suspension of colloidal silica (Ludox HS 40) and viscosity modifier to fill fine pores in the coating layer and the membrane. This silica impregnation is then fired at 500 °C, making the filter end-faces and the entrance regions of

the passageways impermeable. This end coating is very hard and further serves to mechanically protect the ends of the filters.

Performing this coating/impregnation led to significant improvement in filter retention with only minor increase in filter pressure drop, as may be seen in the representative results presented in Table 4. In general, the results reported below are for such "caulked" filters.

Table 4: Improvement in Filter Retention by Coating Ends
(Aqueous Silica Suspension Test)

Membrane Type	No. of Samples	Before Coating		After Coating	
		Retention	Std. Dev.	Retention	Std. Dev.
SiC membranes (w/o frit)	8	87.7%	6.6	97.7%	3.5
Frit-bonded membranes	6	92.3	7.5	98.5	2.9

Results and Discussion: Lab Scale Filters

During this year substantial membrane development was conducted using SiC monoliths with a diameter of 1", a length of 12", and a passageway size of 2 mm. These were coated with a wide range of SiC-based and zircon-based membranes, as outlined above. Regardless of the inorganic membrane forming material, in general, the higher the content of fugitive pore formers in the membrane forming slip, the lower pressure drop and the particle retention. Much of the work this year has focused on this tradeoff to find a suitable compromise between the reduced pressure drop and the reduced retention. Data are presented in Figure 2.
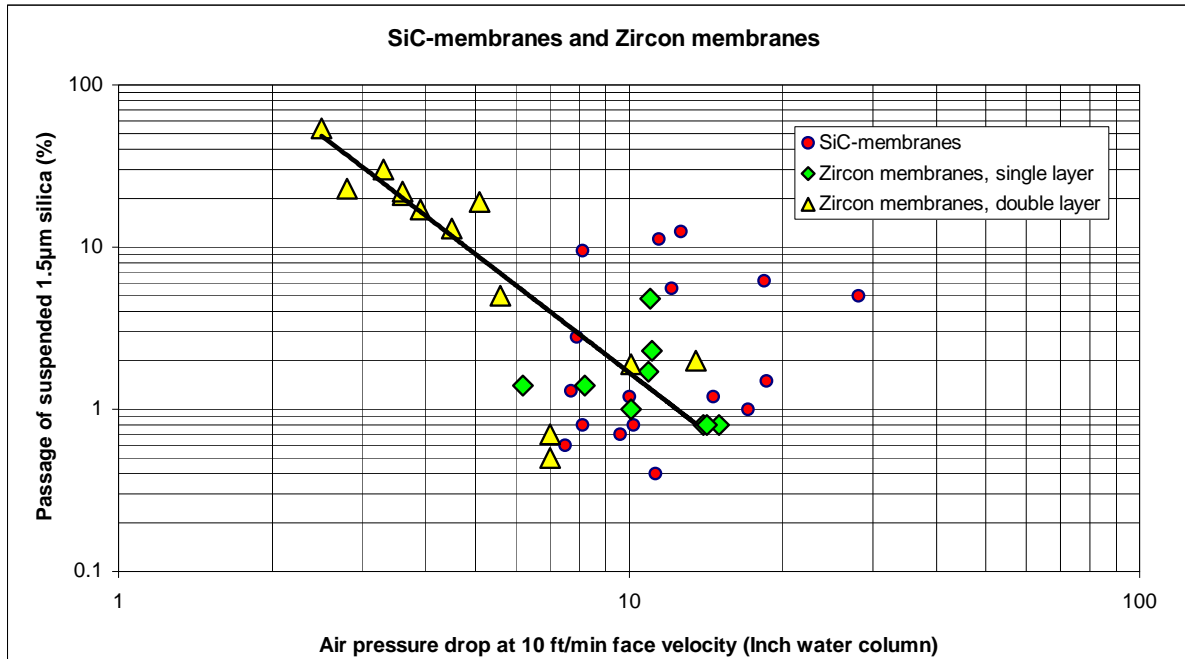

Figure 2. Silica Retention vs. Pressure Drop (@ 10 ft/min RT air flow)

Shown in Figure 2 is the relationship between retention for 1.5 μm silica particles in aqueous suspension as a function of measured pressure drop for air flow through the filters at a filter face velocity of 10 ft/min. Changing from the SiC membrane to the zircon membrane provided much

better control over membrane properties, as can be observed in the lower scatter around the trend line for zircon membranes than was observed for the SiC-membranes (trend line not shown). Going from a single layer zircon-membrane to a composite membrane of more layers made it possible to optimize the membrane properties further, e.g. reduce the layer thickness of the membrane second coat, and therefore even lower pressure drop has been obtained without sacrificing the high retention. The best result obtained was 99.5% silica retention at a pressure drop of 7" H_2O at 10 ft/min face velocity. This retention corresponds to HEPA filter performance, as will be elaborated on further below, when silica retention data are correlated with airborne aerosol HEPA challenge test data.

As an example of an aerosol challenge test, early in the year, one lab scale filter (1" diameter by 12" length), SC-254, was tested at EERC with polydisperse NaCl-vapor in order to measure the retention properties (EERC report in Appendix C). The PM2.5 emissions were fairly constant, 0.0085 mg/m^3, but the clean air flow pressure drop at 19.7 inches of water column @ 10 ft/min excludes this membrane from most applications where low overall pressure drop is required. This was an uncaulked filter, and the constancy of the retention of the filter as it collected aerosol particles (building pressure drop) is characteristic of a filter with a small fixed leak. These data are presented to show this effect. Nevertheless, the retention level is very high relative to that for a non-membrane-coated monolith filter. For the latter, a typical retention level would be only about 50% in this test (85-90% soot retention on diesel exhaust).

Data showing the relationship between CeraMem's aqueous silica retention measurement and HEPA filter aerosol challenge tests are shown in Figure 3. These data were collected on several laboratory filters (1" x 12") coated with both SiC and zircon membranes. This figure shows the PAO and DOP aerosol retention values obtained at SRTC and ATI (Baltimore), respectively, plotted against the silica retention in CeraMem's QC test. Conclusions that can be drawn from the results are the following:

- A reasonable correlation exists between the aerosol challenge tests and CeraMem's liquid filtration QC test.
- Filters can be made which pass the standard ASTM HEPA filter test. To the best of our knowledge, these are the first ceramic filters that have exhibited this level of retention. While the pressure drop is high for diesel applications, it is quite low for a rigid barrier filter with this retention level (much higher than that needed for a diesel filter).

It is necessary to relate these retention results to expected PM2.5 removal efficiencies in actual gas cleaning processes. First, for HEPA filters, by definition a HEPA filter must pass an aerosol challenge test at a retention level of 99.97% for 0.3 μm particles. This is for an "unconditioned" filter. From Figure 4, this retention level would correspond to about 98% retention in the CeraMem silica QC test. From Figure 3, this would correspond to an ambient airflow pressure drop of about 10" H_2O at a face velocity of 10 ft/min. For a rigid barrier filter (as opposed to a Fiberglas® filter) this performance may be considered very good.[1]

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[1] CeraMem is supplying similar filters to DOE's Savannah River Site (SRS) for evaluation as washable HEPA filters used on high level waste tank vents. The response of DOE staff to the performance level of the monolith filters achieved in this SERDP program is highly positive. CeraMem is now making prototype full size filters for the process development group at SRS for development of a back wash process for permanent HEPA filters.
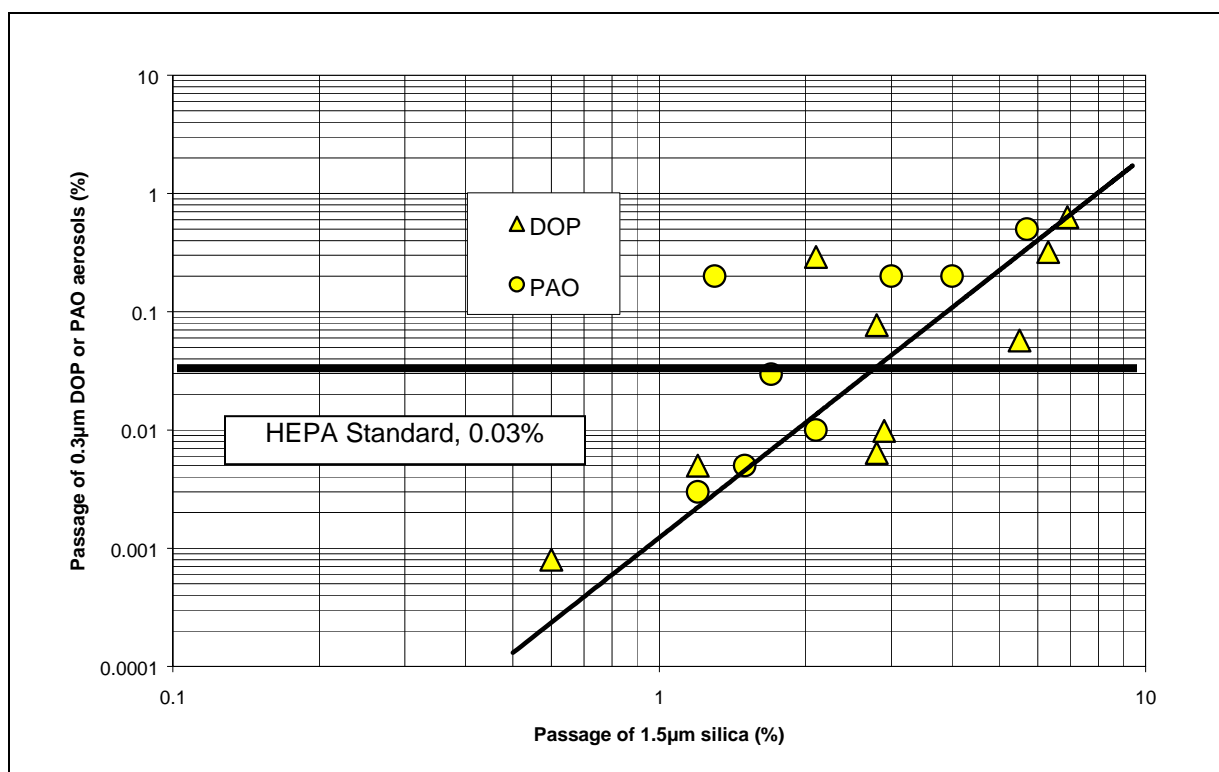
Figure 3: Passage of Airborne 0.3 μm Aerosols vs. 1.5μm Aqueous Silica Particles

For other, less-demanding applications, such as hot gas filters for incinerators or diesel traps, the retention requirements are substantially less demanding. This is due to two factors. First, the removal efficiency required to meet compliance is substantially below the HEPA filter requirement. Second, and quite important, is that the ceramic filters quickly "condition" in the initial filtration process. That is, ash present in the dirty gas irreversibly "caulks" small defects or larger pores. This has been observed previously by CeraMem in several pilot plant trials with earlier cordierite-based filters. Representative data were provided to SERDP as part of the initial project proposal.[2] These data were for repeated NaCl aerosol challenge tests performed on a filter at EERC with intervening backpulse regeneration. After a few cycles, retention of NaCl aerosols at the 0.3 μm size increased from 90% to >99.99% (not measurable). On this basis, we conclude that filters with greater than about 90% retention for aqueous silica (CeraMem's QC test) will condition to be substantially absolute filters for non-HEPA applications. Furthermore, the initial pressure drop for a virgin filter (ca. 5" H_2O, from Figure 2) will increase in an actual application. This will, in part, be due to the conditioning effect, but such increase was found by EERC to be modest. Rather, the sustainable pressure drop of monolith filters in an actual application depends primarily on the effectiveness of the regeneration process. The latter, for incinerator off gas filtration, is ash removal by backpulse regeneration. For diesel filters, regeneration is by *in situ* soot oxidation. However, ash present in diesel exhaust will caulk monolith filters irreversibly. This is especially the case if a fuel borne catalyst is used to assist oxidative regeneration.

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[2] Supplementary technical information in CeraMem Proposal: "Development of a catalyzed ceramic filter for combined PM2.5 removal and VOC and CO oxidation", CPSON 99-01

Based on these results and observations, the following conclusions can be drawn:

- Different ceramic membranes have been developed that are "manufacturable" and which provide the desired high retention efficiency for PM2.5 and low pressure drop.

- Procedures have been developed to deposit and caulk membranes that have HEPA filter performance. These compact filters have a high temperature capability and are regenerable by backwashing.

- The preferred membrane, based on development during 2000, is a glass-frit-bonded zircon membrane deposited on the SiC monolith.

Fabrication Procedures for Full Size Filters

Monoliths selected for prototype full size filters have a diameter of 5.66", a length of 8" to 12", and a passageway size of 0.079" (2 mm). The area of filters made from these monoliths is from about 13 to 20 ft^2, depending on length. These monoliths were developed, and custom fabricated for CeraMem, by the Danish company LiqTech Aps (Lyngby, DK). The target pore size was 15 µm. Pore size and size distribution were measured for samples from full size filter monoliths (ECI, Salt Lake City, UT), and a representative pore size distribution is shown in Figure 4.
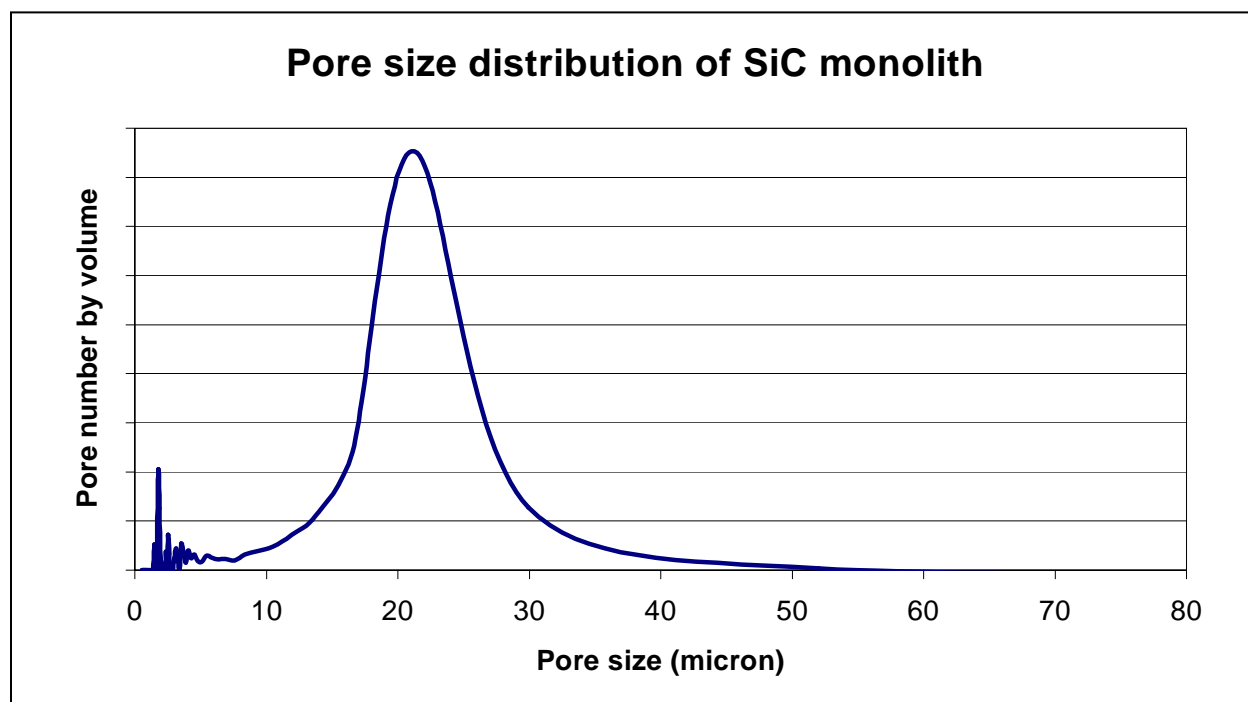

Figure 4. Representative Pore Size Distribution for SiC Monoliths

In general, the control of external dimensions of the monoliths was excellent and they were free from external "skin" defects. However, in early lots of monoliths, internal defects were present, arising from either the extrusion or the drying parts of the production process. Such defects were identified by a CeraMem-developed "graphite test", in which the monolith is temporarily plugged in the checkerboard pattern of Figure 1 and graphite powder is loaded into one end. Any defect in a monolith wall leads to graphite passage into a neighboring passageway. This is observed readily in the graphite pattern on a collection paper at the opposite end as a darkened

spot other than the normal offsetting checkerboard pattern. In general, for any monolith to be used as a filter, a small number of defects in a monolith can be tolerated in that the leaky monolith passageways can be sealed at the ends, removing them as flow paths in the final filter. However, the initial lots of full size monoliths from LiqTech had unacceptable levels of internal defects. CeraMem worked with LiqTech to evaluate the monolith manufacturing processes, making some changes, with the result that filters received by mid-year were of uniformly acceptable quality.

These monoliths, after plugging any cells with internal defects, were used to make prototype full size filters. Mixing of membrane casting slips, as well as coating, drying and firing steps followed the procedures used for laboratory scale filters, with adjustments for increased monolith size. A spin casting system capable of handling monoliths of weight up to 30 lbs was assembled for the coating steps. A full size filter is shown in Figure 5.

Figure 5. Full Size Filter (5.66" diameter x 12" length, 20 ft^2 area)

Testing of finished filters included in-house testing of airflow pressure drop and retention of suspended silica. Both testing methods were performed in scaled up versions of the equipment used for similar testing of the smaller laboratory scale pieces. In addition, selected filters were tested at ATI's ORNL HEPA Filter Test Facility by a standard method, IAW Mil-STD-282. The filters were tested by ATI at face velocities 6 ft/min and 1.2 ft/min. (ATI report in Appendix B).
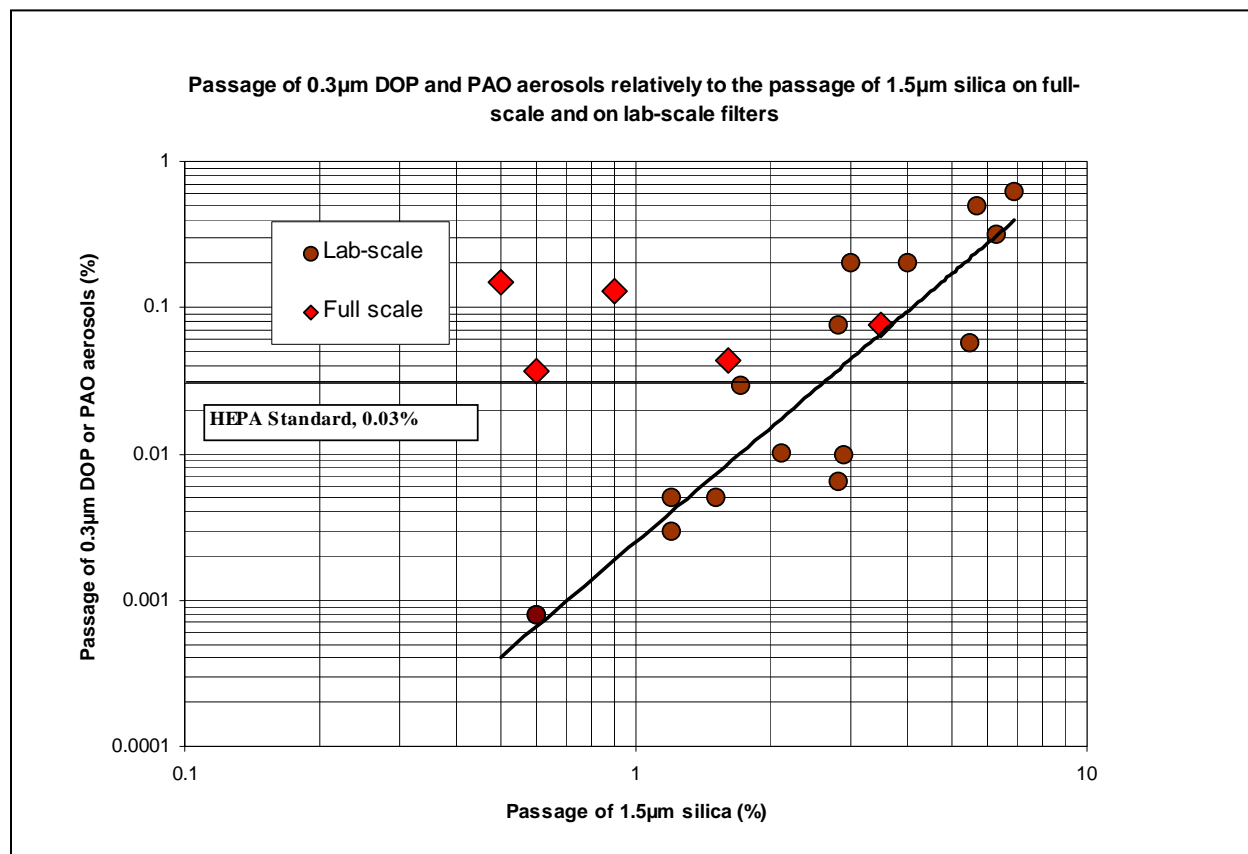
Full Size Filter Test Results

In general, full size filters showed comparable characteristics to those of the lab scale filters, as summarized in Table 5. Scaling up did not significantly change the performance of the filters. The retention range of 84-98% on filters with relatively narrow range of pressure drop indicates that membrane defects were present in some cases. Some of these low-retention membranes have been caulked (as described above) resulting in a significant increase in retention.

Table 5: Comparison of Properties of Lab Scale and Full Size Filters

Filter Characteristics (all 12" long)	Air flow pressure drop @ 10 ft/min	Retention of Suspended Silica
SiC membranes (4/4), 5.66" diameter filter	19-22 "wc	86-98 %
SiC membranes (4/4), 1" diameter filter	14-20 "wc	84-99 %
Zircon membranes (2/2), 5.66" diameter filter	14-18 "wc	91-98 %
Zircon membranes (2/2), 1" diameter filter	12-15 "wc	86-98 %

ATI has tested five full size filters at ORNL in a HEPA filter test with DOP aerosol. Results, shown in Figure 6, do not fall onto the same trendline as the results from laboratory scale filters, also shown in Figure 5 (from Figure 3).

**Figure 5: Retention Data for Full Size and Laboratory Scale Filters
Correlation of HEPA Test Results with Aqueous Silica Retention**

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Since these results were obtained just prior to preparation of this report, is not clear if this is due to the use of different test-stands, testing methods, issues related to the different housings and sealing principles, or another factor. This discrepancy is being resolved at the moment with retests both at CeraMem and ATI. In any event, two of the filters were within 1% of the HEPA test standard and two other were fairly close. The levels of retention are still very high and are judged to be more than sufficient to meet the needs for PM2.5 removal from DoD sources.

FY00: Task 7: Develop and Test Full-Size, Passively Regenerated Catalytic Filters

The intended application for filters to be developed in this task is diesel exhaust aftertreatment. Based on discussions with the TAC, confirmed by a presentation at the fall SERDP Conference (Peter Schill, US TACOM RD&E Center) and subsequent discussions, the following conclusions were reached.

- DoD diesel sources will use intermediate- to high-sulfur fuels. While the consumer market for diesels can anticipate low-sulfur fuels and use control devices that require such fuels, emissions control for military sources must plan on high-sulfur fuels.

- Catalyzed diesel traps will almost certainly require a precious metal catalyst to achieve a significant reduction in regeneration temperature from the thermal regeneration of non-catalyzed filters (*e.g.*, reduction from 600°C to 400°C). Unfortunately these catalysts increase the production of sulfate due to the facile oxidation of SO_2 to SO_3. Also, catalyst poisoning by sulfur is an issue.

As an alternative to catalyzed filters, the use of a fuel borne catalyst (FBC) is being actively pursued by several organizations. Peugeot is commercially implementing passive regeneration of their on-vehicle filters using an FBC-based strategy. Most of the FBC's are metal oxides that do not as readily oxidize SO_2 and are more resistant to sulfur poisoning. Further, they provide a substantial reduction in soot oxidation temperature from that achievable with the best-catalyzed traps (*ca*. 300°C vs. 400+°C). Part of the experimental program will be directed at measuring the impact of fuel sulfur concentration on catalyst activity and on sulfate formation.

At the recommendation of the TAC this task, which originally included the short term testing of a genset in CeraMem's parking lot, will be changed. In lieu of the genset test, much more extensive testing on a 1.9 liter diesel engine will be performed at Penn State University (PSU) in the Energy Institute under the direction of Professor André Boehman. These tests will be on a well-characterized engine, processing full exhaust flow, evaluating different regeneration schemes, with and without fuel borne catalysts. We intend to procure from Engine Control Systems (ECS) a standard diesel exhaust aftertreatment system to house our filters. We should have the tests underway in the second quarter and will continue, we anticipate, throughout much of the year. We are now defining with Professor Boehman a detailed scope of work and associated budget for the testing. The current iteration is included as Appendix D.

We anticipate that these extensive tests will substitute for the short-term tests planned in this task and the field tests originally planned as Task 11 in FY 2001.

FY00 Task 8: Develop and Test Full-Size, Backpulse-Regenerated Catalytic Filter for VOC Oxidation.

Based on an analysis of the target application (incinerator off gas) need, we no longer intend to use a catalyzed filter. This is due, in large part, to the dioxins emission issue raised by the TAC. To address this issue, we will use the CeraMem filter at an intermediate temperature (*ca*.

500 °C). Removal of ash at this temperature avoids the well-documented formation of dioxins in the presence of ash in the temperature range of 250-400+ °C (*e.g*., Chapter 3: Incineration. Committee on Shipboard Pollution Control, Naval Studies Board, National Research Council[1]). Catalytic oxidation would be performed at a somewhat lower temperature (limited by catalyst stability) to remove gas-phase dioxins formed during combustion and not removed by the filter, hydrocarbon vapors, and CO.

Thus, the process approach is to filter the incinerator off gas hot, followed by oxidation over a monolith-supported oxidation catalyst, followed by cooling with dilution air and charcoal adsorption (if needed) to remove any remaining toxic materials (e.g., Hg).

In lieu of the short-term tests originally planned at CeraMem, the TAC and CeraMem concur that tests under more controlled conditions, for longer term, and with dioxin monitoring are preferred. We are now evaluating several potential locations for these, and expect to choose one by the end of March. The sites are PSU, the Energy and Environmental Research Center at the University of North Dakota (UNDEERC) in Grand Forks, ND, MSE Technology Applications, Inc. (MSE) in Butte, MT, and two medical waste incinerators in New England. The first three have very well equipped and instrumented pilot combustion facilities or incinerators. The local medical waste incinerators will require more supporting sampling and instrumentation capabilities from a sampling/analytical laboratory source. Multiple oxidation catalyst samples, intended for dioxin oxidation, will be provided by Prototech Corporation.

It is our anticipation that these longer-term, larger scale trials will begin in the summer of FY 2000. It is our expectation that these will fulfill the requirements of this task and Task 11 of FY 2001. A preliminary description of the testing is included in Appendix E. One issue is the question of the capability to perform dioxin testing for the generation/destruction of dioxins within the existing budget. A second question relates to the political acceptance for injection of dioxin standards for measuring fate and effects.

5. SUMMMARY AND CONCLUSIONS

a) The capability of making full size filters with a range of retention characteristics and corresponding pressure drop has been established.

- Filters with high retention (expected to be >99% for soot) and relatively low pressure drop can be produced for diesel exhaust aftertreatment.

- Filters with very high retention and moderate pressure drop can be produced for very high PM2.5 removal requirements.

- Filters with HEPA filter performance can be produced with high, but acceptable, pressure drop for DoD applications requiring permanent, regenerable HEPA filters.

b) For diesel exhaust applications, based on the need for handling moderate-to-high-sulfur fuel, the concept of a catalyzed diesel trap has been dropped due to the limitations of either (a) catalyst poisoning by sulfur, or (b) catalytic oxidation of SO_2 to sulfate. Filter regeneration will be either by non-catalyzed thermal oxidation or oxidation assisted by fuel-borne non-precious metal oxidation catalysts.

c) For point source applications, such as incinerators, the addition of a gas phase oxidation

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[1] URL: http://books.nap.edu/html/ shipboard_pollution/chapter3.html

catalyst to the filter to remove VOC's and CO (also dioxins) has been dropped in favor of adding the oxidation catalyst as a separate unit directly downstream of the PM2.5 filter. This allows much greater flexibility in terms of catalyst selection and independent sizing of the catalyst device.

d) The economic evaluation of the technology demonstrates that the technology is economically viable for DoD use.

e) The ceramic-membrane-coated, monolith filters are developed to the state where meaningful field trials are the next step in product and process development. Two significant trials will take place during 2001.

APPENDIX A

Economic Feasibility Analysis Report

For

Development of a Catalyzed Ceramic Filter for Combined Hot Gas PM2.5 Removal and VOC/CO Oxidation

CP1120

Bruce Bishop and Robert Goldsmith

CeraMem Corporation
12 Clematis Avenue
Waltham, MA 02453

October 31, 2000

1.0   Introduction

The Department of Defense (DoD) has a large number of particulate emissions sources that may be impacted by new PM2.5 regulations. The USEPA has proposed new PM2.5 regulations in addition to those already enacted to control PM10 because it believes that current standards do not sufficiently protect the public health. The proposed new standard presents a clear challenge to many industrial and Governmental segments of our society. Indeed, it is questionable whether technology exists to control PM2.5 economically from the majority of sources.

The Defense Department, through the Strategic Environmental Research and Development Program (SERDP), has identified a number of emissions sources that need enhanced control systems if proposed regulations become reality. These sources include engine test cells, diesel power generation sets (gensets), shipboard diesel engines, heavy-duty mobile diesel sources, trash incinerators, medical waste incinerators, demil waste incinerators, and steam boilers. These sources are emitters of fine particulate matter, potentially subject to the new PM2.5 standard, and many are emitters of significant amounts of volatile organic compounds (VOCs) and carbon monoxide (CO).

The focus of the present project is the development of a novel technology for PM2.5 removal with substantially 100% removal efficiency. The technology is a highly compact, ceramic-membrane-coated monolith filter. Moreover, this same filter can be coated with non-selective oxidation catalysts for the combined, simultaneous removal of particulate and/or vapor-phase organics and carbon monoxide. While no single emission control technology will address all of DoD's needs in this area, this catalyzed filtration technology may be able to address specific needs.

1.1. Technology Description

The base technology for the products to be developed is shown in Figure 1. The mechanical structure is a porous, ceramic honeycomb monolith, similar to those used in automotive catalytic converters. The material can be of cordierite ($2MgO-2Al_2O_3-5SiO_2$) or silicon carbide (SiC). The monoliths can have 64 to 100 square passageways per square inch (cpsi) of frontal area that extend in parallel from the inlet end face to the outlet end face. As a diesel particulate trap, the porous honeycomb has its inlet and outlet passageways plugged in a checkerboard pattern. Gas flow through the filter is constrained to pass through the wall structure of the monolith. The soot in the gas stream is removed both on the wall surface and throughout the thickness of the porous wall. Particulate removal efficiency is typically 80-90%. Cleaning in order to restore or nearly restore the original gas flow resistance of the filter is by thermal or catalytic oxidation. Backpulsing of the filter with gas or liquid to remove the particulate is hindered due to the trapping of significant amounts of material within the pore structure.

A modification to this standard trap is highlighted in Figure 1. A porous ceramic membrane coating on at least in the inlet passageways of the monolith will retain the particulate on its surface. If the membrane coating is thin, it can have a much smaller pore size than that of the monolith support increasing the particulate removal efficiency many orders of magnitude without added undue gas flow resistance to the filter. Furthermore, with the particulate on the surface of the membrane, backpulse regeneration of the filter is much easier since the particles do not have to be removed from the depth of the pore structure. Consequently, the addition of the membrane coating transforms the trap into a high-efficiency particulate filter with enhanced regeneration capability.

Figure 1. CeraMem™ gas particulate filter schematic
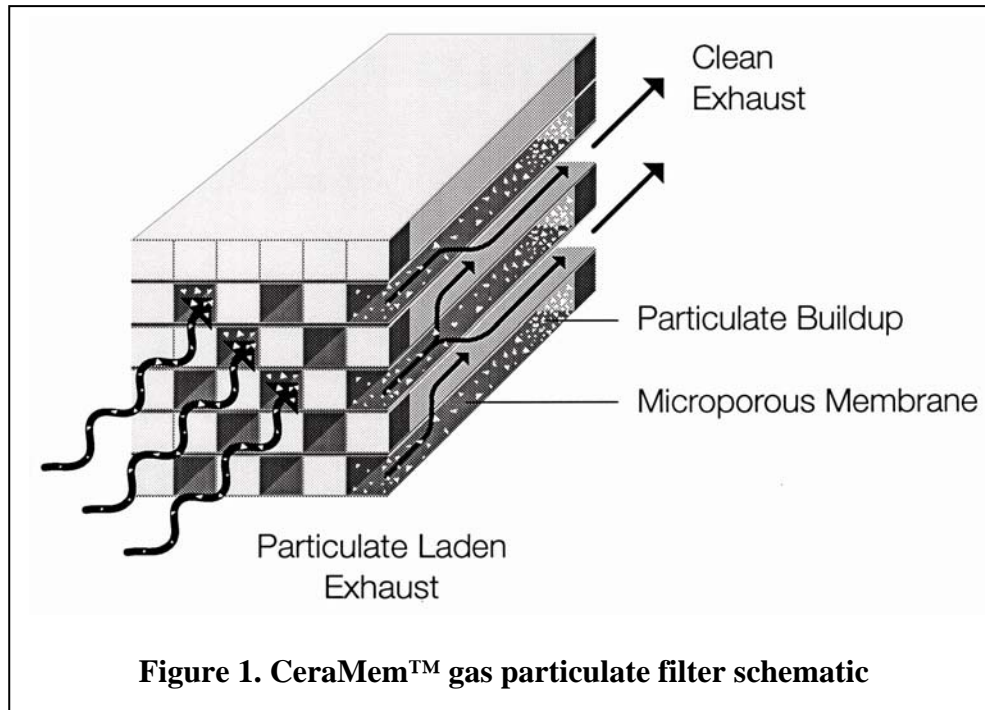
The base filter technology can be augmented by the strategic addition of catalyst within the filter structure. The types of catalysts to be evaluated in this SERDP program are oxidation catalysts, for either soot particles or VOC/CO gases. An outline of potential filter modifications is shown in Figure 2. These potential products can cover many of the DoD needs in air-borne emissions.

Figure 2. Products to be developed in SERDP Project CP1120

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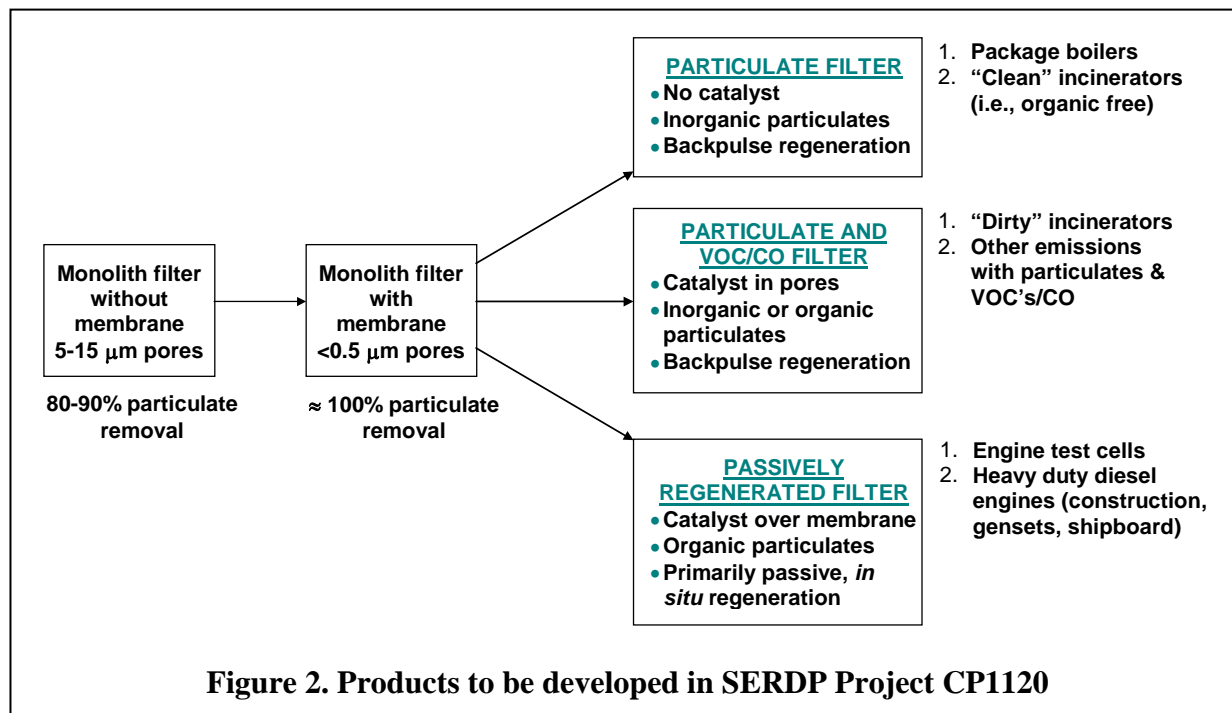
1.2. Purpose of Analysis

In this early stage of development, a preliminary analysis of the feasibility of the proposed products was conducted in order to ensure that process economics were realistic and no development efforts were wasted on unproductive paths. The following information describes the estimated prices of each filter type, systems in which the filters would operate, and the impact of these prices on some selected applications.

2.0 Filter Price

The filter price model was developed based on a number of factors. The cost of monoliths were based on filter volume and historical manufacturing costs in $/in^3 extracted from CeraMem's experience with cordierite (Corning and NGK) and SiC (Stobbe Tech). Membrane coating and filter canning (i.e., mounting of ceramic filters into metal cans in order to attach them to filtration systems) costs were similarly based on CeraMem experience in a commercial filter business. This experience comes as a result of operating CeraMem Separations, Inc., which was a ceramic filter spin-off company that included Exxon Chemical Company and Corning Inc. as investors. This company was sold to Corning, Inc. in 1997. Manufacturing yield in this model was set at 90%. Catalyst coating cost was supplied by Prototech Company, a division of United Catalysts, based on noble metal prices as of May 2000. Prices were extrapolated from filter costs, other than catalyst coating that was passed through without additional mark-up, using gross margins typical in filter businesses. Prices were then normalized to filtration area of each filter configuration, calculated from diameter, length, and cell density.

The results of the model are summarized in Figures 3-6. Figure 3 shows the effects of filter size and manufacturing volume on price. Figure 4 indicates the effect of membrane type along with filter size on price at a manufacturing volume of 100 filters per week. Figure 5 reveals how monolith material can affect filter price for each of three different filter sizes, once again at high manufacturing volume. Figure 6 compares prices for each different product type using either cordierite or SiC monolith supports and assuming large filter size and manufacturing volume.

Based on this model, the following observations can be made. The type of membrane deposited, either one or two layers, does not have a strong effect on price. This is because coating costs are relatively small compared to monolith manufacturing and about the same as filter canning. Filter size and manufacturing volume affect filter price substantially. These effects are not surprising given the economies of scale. The time to produce small or large diameter filters is essentially the same and hence when the price is normalized to unit area, the higher surface area filter will have the lower normalized price. Similarly, for increased manufacturing volume, indirect costs can be spread over a much larger number of units thereby reducing cost per unit. In addition, efficiency tends to increase with manufacturing volume also reducing cost and, in turn, price. The largest impacts on price are from monolith material and catalyst addition. Cordierite can be made much less expensively than SiC. The raw materials, die wear, and firing costs are substantially less expensive for cordierite than SiC. However, other attributes of SiC monoliths such as lower pressure drop, more uniform pore structure (leading to better membrane coatings), greater structural integrity, and potentially better thermal shock resistance may result in more attractive life-cycle costs for a filtration process. The addition of noble metal catalysts for either soot or VOC/CO oxidation will clearly increase price due to the cost of such materials. An objective of the program should be to minimize the quantity of noble metals necessary for catalytic oxidation.
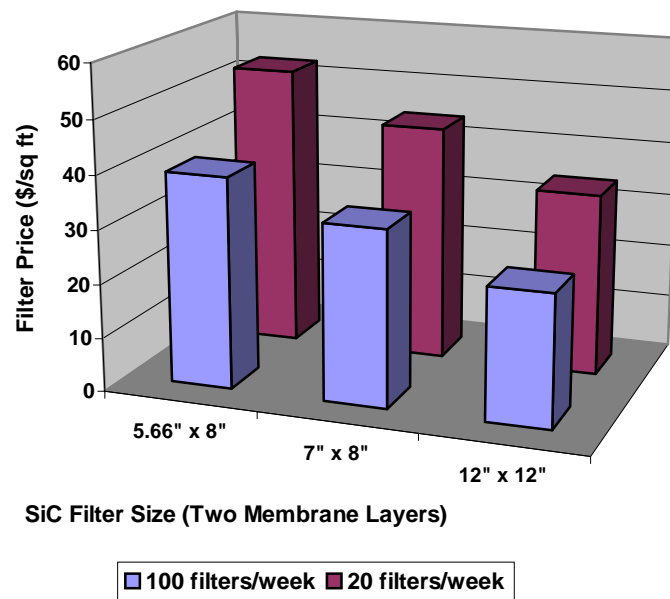
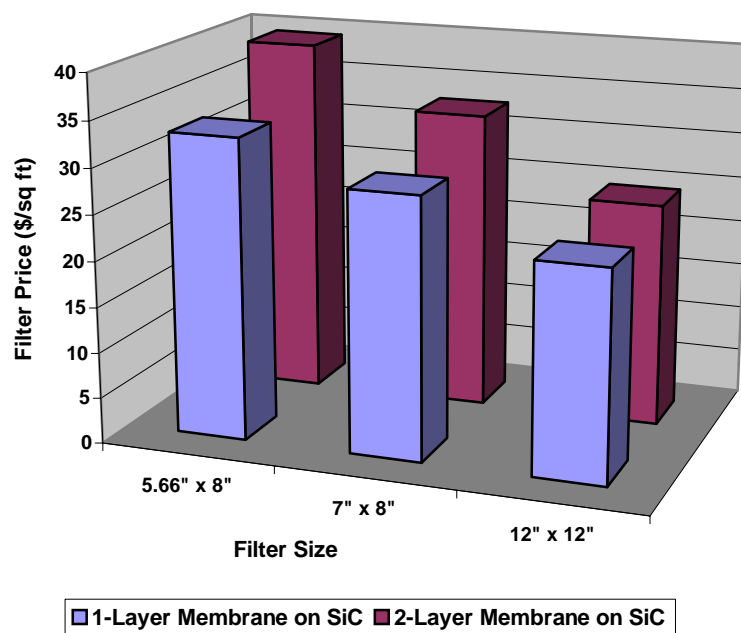**Figure 3. Effects of size and manufacturing volume on price of SiC filters**

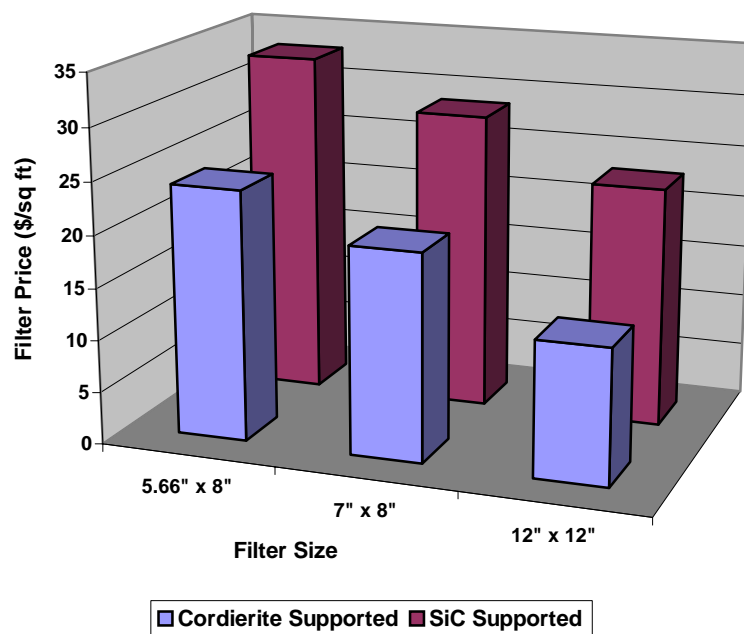**Figure 4. Effects of size and membrane type on price of membrane-coated SiC filters**

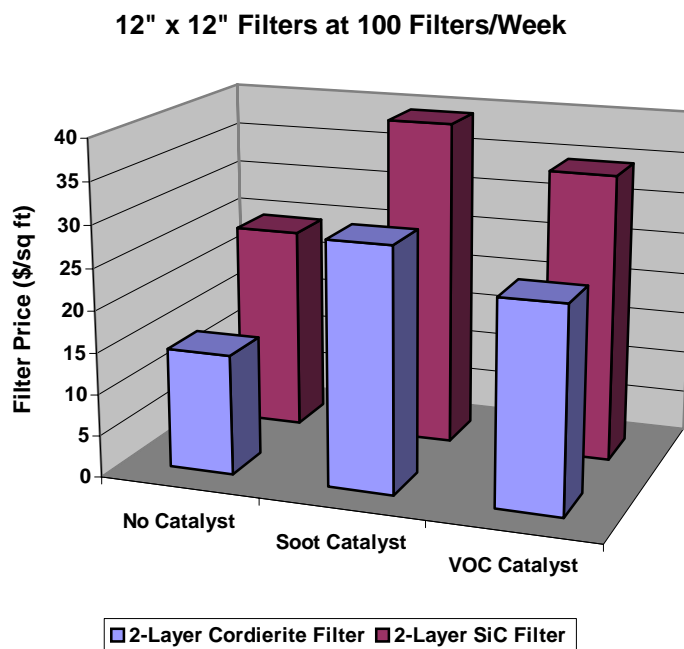**Figure 5. Effects of size and monolith material on price of membrane-coated filters**

Figure 6. Effects of monolith material and product type on price of filters

3.0   System Price

Once the filters have been fabricated, they must be installed in a system in order to operate them in a filtration process. Many of the DoD applications of interest will require filtration systems similar to those used with fabric bag filters or ceramic candle filters. Based on system quotations in the industrial and utility markets that CeraMem Separations Inc. had prepared as part of its business, CeraMem Corporation has prepared system prices as a function of filtration area.

Figures 7 and 8 present the price of filters, system hardware, and the total system for membrane-coated SiC filters without and with oxidation catalyst. For non-catalytic filters, the filter price runs about 25% to 35% of the total system price, which is fairly typical of industrial hot gas filtration systems. For these systems, the price in $/ft^2 ranges from about 200 to 70 as the system size increases. For cordierite-based filters (not shown), this range is about 160 to 55 as a result of the lower monolith materials cost. This initial price differential may be compensated for by lower life cycle costs (e.g., longer filter life) or performance (e.g., higher filtration efficiency) of the SiC filters but is yet to be proven.

For catalytic filters, the filter price ranges from about 25% to 50% of the total system price. A noticeable difference in filter pricing is that the cost of noble metal catalyst appears to minimize the reduction of price with increasing volume. Assuming that material costs dominate the catalyst coating cost, this effect is logical in that cost will be directly proportional to size. This makes filter price more comparable to system hardware and hence a larger percentage of the total system price. At small system size, the normalized system price does not increase substantially due to the addition of catalyst to the filters; however, for larger systems the price increases to about $80/ft^2 for SiC filters and $70/ft^2 for cordierite. The range of system price shown here is typical of industrial filtration equipment of about $100/ft^2 and shows that the products to be developed are economically feasible in industrial-type systems.

Figure 7. Non-catalyzed SiC filters, hardware, and system price versus filtration area
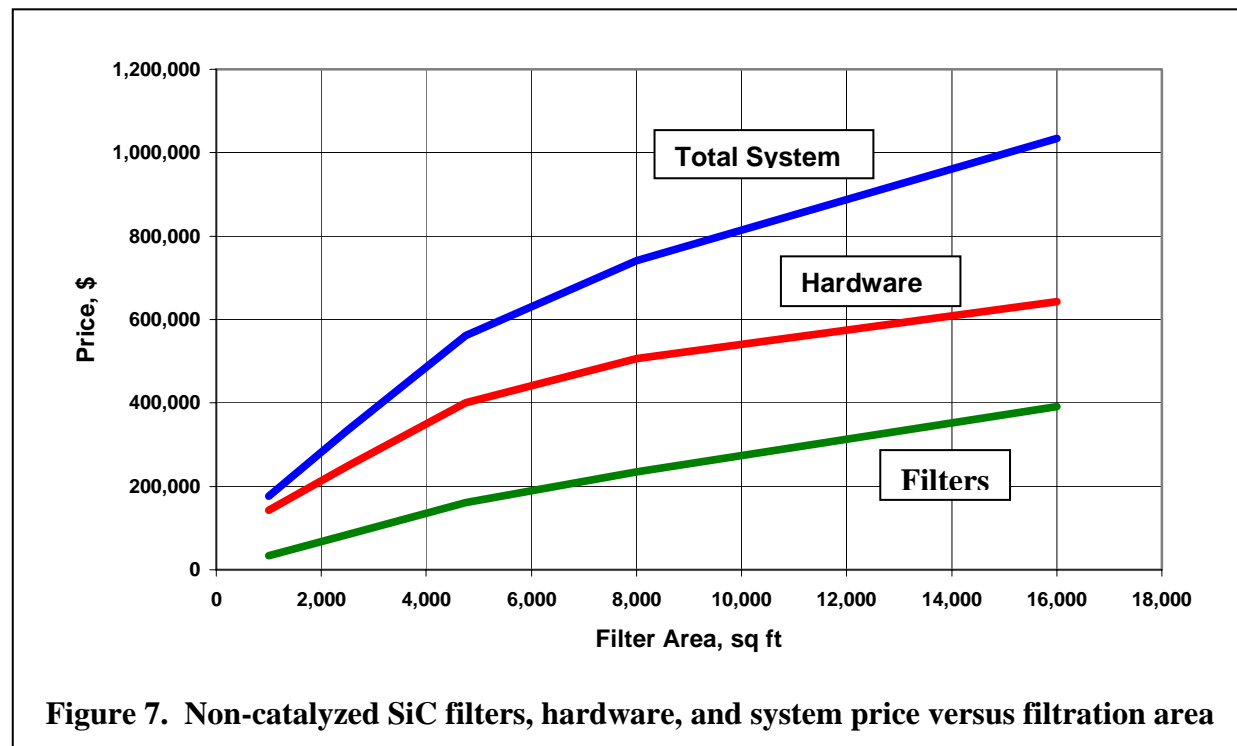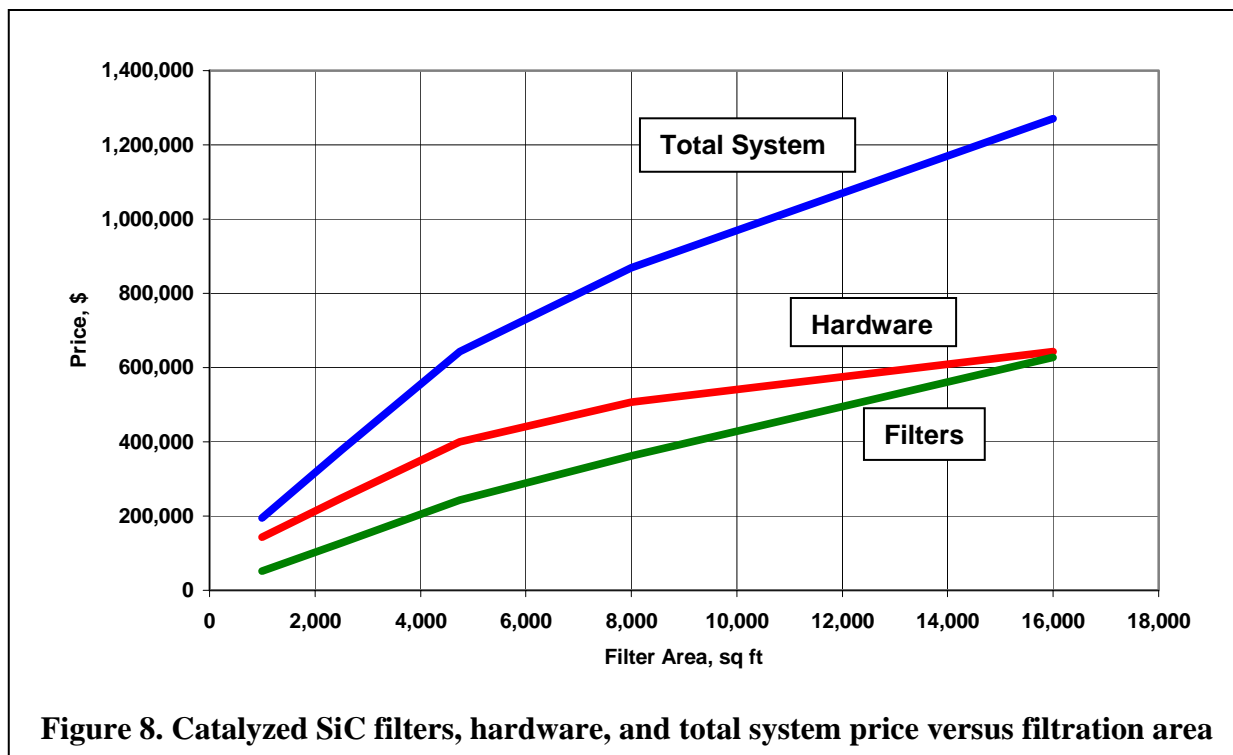
Figure 8. Catalyzed SiC filters, hardware, and total system price versus filtration area

4.0   Potential Applications of Interest

DoD has a number of emissions sources that must be treated if new clean air regulations are implemented. These sources, along with some duel-use applications, include the following:

1. Jet engine test cell (JETC) exhaust,**

2. Diesel engine exhaust including

    a. Stationary sources such as portable electrical generator sets** and on-board marine diesels, and

    b. Mobile sources such as trucks and off-road equipment,

3. Incinerator off-gas from

    a. Naval vessels (e.g., aircraft carriers),

    b. Cruise line ships, and

    c. Military medical waste incinerators,

4. Packaged boiler flue gas,

5. Two-stoke engine exhaust, and

6. Exhaust gas analyzer test gas.**

Those applications marked with ** have been further analyzed and the results are discussed below.

4.1 Jet Engine Test Cells

Of all the potential applications, JETCs are perhaps the most difficult application to address for air emissions reductions. Due to the volume of gas generated, including augmentation air, and the force with which the gas flows, setting up a filtration system to meet DoD's needs is a challenge. Some of the issues that were considered include

1. Placement of hardware inside the tunnel that may be damaged by the gas jet,

2. Decreasing the sound suppression effectiveness of the tunnel,

3. Meeting the structure height limitations near the flight line,

4. Disqualification of the engine test due to changes in system backpressure (or any other attributes), and

5. Capital and operating costs.

As shown in the system schematics below, the jet engine test stand is located in front of the tunnel. Exhaust gas from the engine is extracted from locations along the length of the tunnel and drawn into the filtration system that removes particulate matter and gas phase pollutants. The system can be described in three different sections: augmentation air inlet, exhaust gas tunnel, and filtration system.

The inlet section to the tunnel can be seen in plan view in Figure 9. The augmentation air is constrained to flow only through the engine so as to keep the engine cool and to minimize the total exhaust gas flow. This minimizes the filtration system size and price. A second benefit is that the exhaust gas temperature in the tunnel is higher than usual. If the gas temperature gets hot enough, soot and gas-phase pollutants will be oxidized in the filters eliminating the need for in-line gas heaters.

Because of the higher temperature, the inside of the tunnel is modified to withstand the hotter conditions, at least at the inlet. As it flows down the length of the tunnel, gas is extracted from two and possibly more locations. It will not be economical to extract all the gas, and in any case, doing so would certainly disqualify the test. However, if enough gas can be extracted to drive the total emissions down below regulatory limits, then the approach might be successful. The gas is drawn through insulated ducts that penetrate the tunnel in three spots at each extraction location (Figure 10).

The gas is directed to the filtration system that consists of two banks of filters, each with 200 filters. The inlet and outlet plenums are located between the two banks of filters. Not shown in this schematic is the tapering of the inlet and outlet plenums to ensure uniform gas flow into and across the filter banks. Each filter is equipped with a backpulse nozzle and venturi to be used to regenerate the filters in case of catalyst failure or if inorganic particles blind the filters. Below the filter tubesheet is located the dirty side of the system where the particulate matter disengages from the backpulse gas and is collected at the bottom of the hoppers. Rotary lock valves can be used to remove solids from the system as necessary. On the clean side of the system, a large high-temperature blower is used to induce gas flow from the tunnel and through the filtration system. The height of the system was intentionally kept low so as to not exceed flight line requirements.
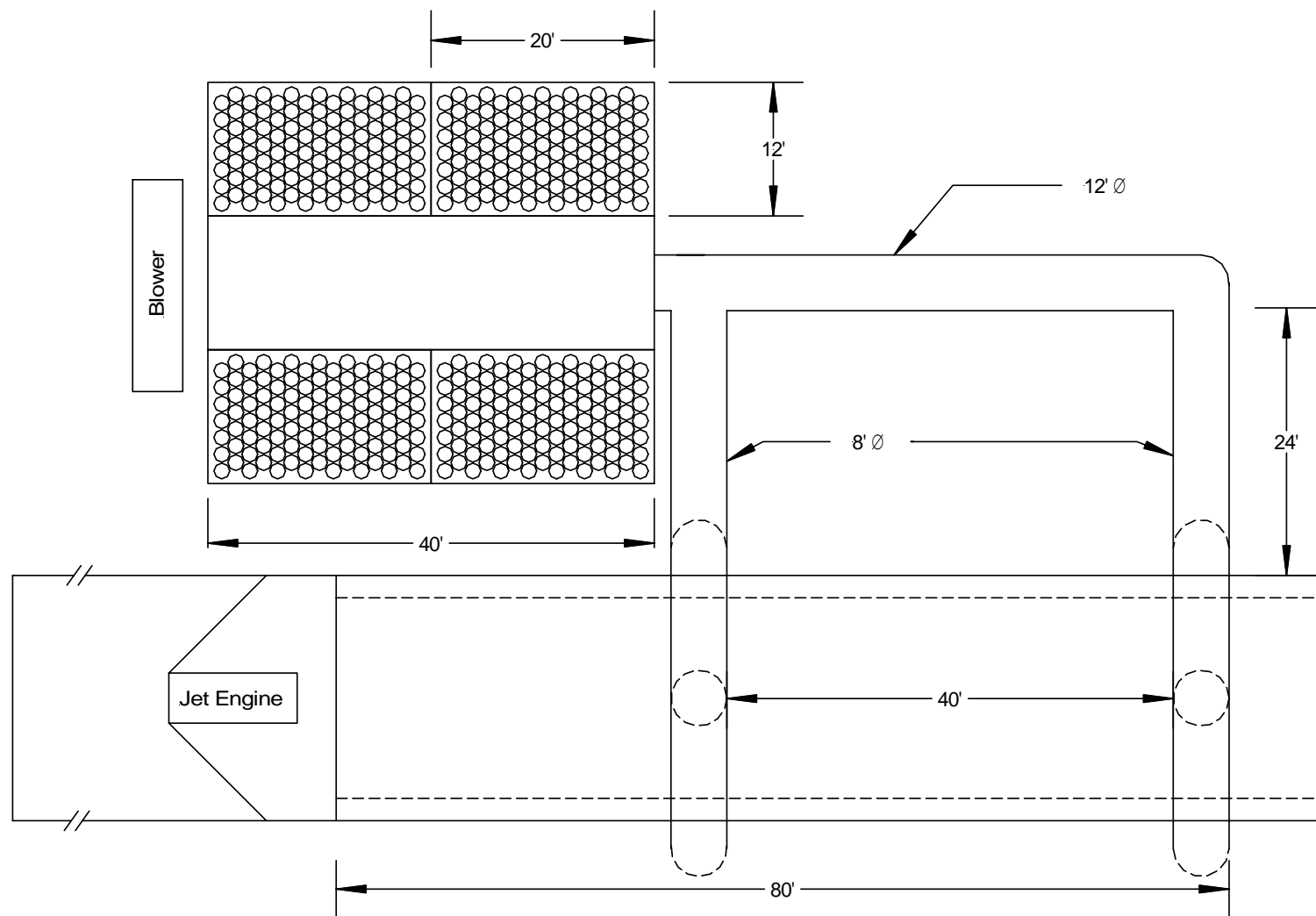
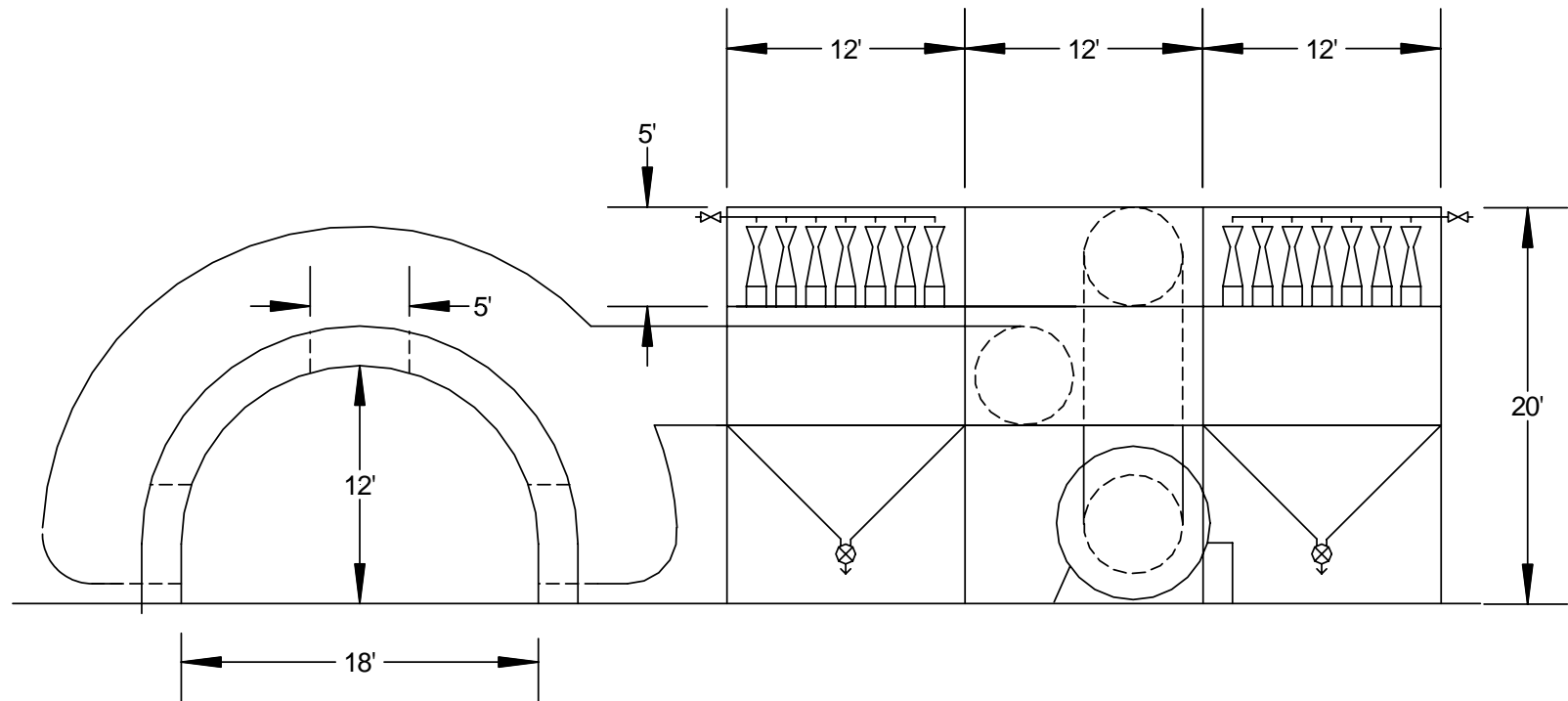**Figure 9. Plan view of JETC aftertreatment system**

Figure 10. End view of JETC aftertreatment system

The life-cycle costs for the JETC have been estimated on the above system approach and the following bases:

- 200,000 cfm each for engine exhaust and augmentation air,
- 9.5 ft/min face velocity through the filters,
- 10 year system lifetime and 5 year filter lifetime,
- 1,000 hours of operation per year, and
- 10 tons/yr of PM and 12 tons/yr of VOC (per SERDP Statement of Need).

Several sources were used to estimate costs for the system. The filters and filtration system were based on costs described earlier in this report. Cell modifications were based on high temperature alloy material costs (Alloy 253 MA, Rolled Alloys, Temperance, MI) and fabrication and construction discussions with hardware fabricators (B.E. Peterson, Avon, MA and Foster-Wheeler, Clinton, NJ). The cost of the high temperature blower was a budget estimate by New York Blower.

The capital, operating, and annualized life-cycle costs are given in Table 1. The outcome of this preliminary analysis is that the cost of removing particulate matter and VOCs from JETC exhaust gas is probably not economic. The cost per ton of pollutant of almost $32,000 is in the same range as that of NOx that Wander and Canfield found with a sorbent system for JETCs.[3] Costs of the NOx sorbent process were found unattractive at $25,000 to $50,000 per ton.

Table 1. JETC Life-cycle Costs

Item	Comment	Cost, $
Installed System Cost		
Filters	400 Filters, 105 ft^2/Filter	$1,500,000
System and Blower	1600 hp Blower	$1,500,000
Test Cell Modifications	Ducting and Tunnel	$350,000
Yearly Operating and Maintenance (O&M) Costs		
Maintenance	3% of Capital	$100,500
Filter Replacement	5 Year Filter Life	$150,000
Power	1600 hp at $0.08/kW-hr	$96,000
Decommissioning		
Cost	One Time Charge	$175,000
Annualized Cost Over System Lifetime		
System and Decommissioning		$352,500
Yearly O&M		$346,500
Total Annual Cost		$699,000
Annualized Cost per Ton of PM and VOC		
Cost		$31,800

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[3] J.D. Wander and C.A. Canfield, "Jet Engine Test Facility Exhaust Emission Control System", JETC97-070798, 1997.

4.2   Diesel Exhaust Aftertreatment

One of the most promising applications for the products to be developed in this program is diesel exhaust aftertreatment. DoD has a large number of diesel-powered vehicles, ships, portable generator sets, and other pieces of equipment. These are coming under closer scrutiny as diesel exhaust emissions are being reduced through regulatory requirements.

In general, diesel sources are either mobile or stationary. Sources such as trucks, tanks, and off-road construction equipment are considered mobile while generator sets and engine test stands are considered stationary sources. The reason for the differentiation has as much to due with the duty cycle as the mobility of the source. Mobile sources have large transients in power output, exhaust flow rate, and pollutant emissions inherent in their duty cycles while stationary sources are much more steady. This makes adaptation of aftertreatment systems to stationary sources much easier than to mobile sources. For this program, it is likely that initial field trials will be conducted using a stationary source of diesel exhaust. Consequently, the preliminary feasibility analysis was performed for such a source.

The selected source was the Air Force's A/M-32-A-86 portable generator set. It has a rated power output of about 100 kW and flow rate of 350 cfm. It was assumed that the maximum allowable backpressure on the engine was 2" Hg. This resulted in a design face velocity of 5 ft/min. In order to keep the system simple, a single filter with about 70 ft^2 of filtration area was selected for costing. This filter would be 10" in diameter and 10" long with 100 cpsi.

Aftertreatment systems in which diesel filters are utilized can be rather complex in their design and operation. It is beyond the scope of this program and CeraMem's capabilities to design such a system for the "-86" generator set. Another approach was taken to determine economic feasibility. The products that CeraMem wishes to develop in this program would directly replace existing diesel traps. The potential for higher particulate removal efficiency, improved passive regeneration, and longer life at comparable operating pressure drop (lower life-cycle costs) would make them attractive if the initial cost was about the same or less than present technology. Hence, this analysis will compare the estimated initial costs of a generalized aftertreatment system with alternative filtration technologies.

Table 2 compares the difference in cost for a series of diesel traps and the resultant change in system cost. It is assumed that the filter canning, catalyst, and system costs would be the same for the same throughput-rated filter regardless of type. The first line is the estimated price for a small, 5.66" diameter, 6" long, diesel trap based on CeraMem's price model. The estimated price is compared to the actual price of the part as purchased by CeraMem from Corning, Inc. This benchmark indicates that the price model is at least realistic. Prices were also estimated for a non-membrane-coated cordierite trap, a membrane-coated cordierite trap, and a membrane-coated SiC trap. As noted earlier in the report, the effect of membrane coating on filter price is not nearly as great as that for monolith material. The SiC filter is much more expensive than the comparable cordierite version. However, when included in the price of a system, the incremental increase in price is only modest. Therefore, given the uncertainty of the estimates, the increase in system price may only be slight to modest. This indicates that membrane-coated SiC traps might be economically feasible if their superior performance can be proven.

Table 2. Diesel Cost Analysis: Incremental Filter Price Comparison

Assumptions: System, catalyst, and filter canning prices are the same for each filter type.

Filter Type	Estimated Price	Actual Price	Incremental Increase*	% Change in System Price**
5.66" x 6" cordierite, no membrane	$140	$125	---	---
10" x 10" cordierite, no membrane	$375	---	---	0
10" x 10" cordierite with membrane	$553	---	$178	2.4
10" x 10" SiC with membrane	$1,396	---	$1,021	14.0

* This is the price increase as compared to a 10" x 10" cordierite trap with no membrane coating.

** System price for 100 kW engine is about $7,300 [Selection Criteria for Diesel Particulate Trap Systems; VERT Experience, www.dieselnet.com, December, 1998].

4.3   Diesel Exhaust Gas Analyzer

The last application to be evaluated is the pretreatment of diesel exhaust prior to entering engine exhaust gas analysis equipment.[4] Discussions with a major analytical equipment manufacturer have revealed a potential need. Engine exhaust gas is filtered at 190°C to remove particulate from gases prior to entering sensitive analytical equipment. Presently, filter housings about 7" long and costing up to $800 with replaceable fibrous filter inserts are used. The inserts need to be replaced every 2 to 8 hours of testing at $10 each. Given that the analytical instruments are run about 2000 hr/yr, the replacement filter cost is about $2,500 to $10,000/yr not including labor costs for replacement. A regenerable filter may be a cost effective substitute for the present approach.

The proposed regenerable filter approach is shown in Figure 11. A small filter coupon, about 1" in diameter and 2" long, is housed in a high temperature fixture. The coupon has heating elements wrapped around it with external temperature control. The fixture has inlet and outlet air purge lines with solenoid valves to control the flow. Particulate-laden gas enters the filter assembly at 190°C after the filter is preheated to keep the gas stream from being cooled. As clean, hot gas flows through the filter to the gas analyzer, particulate is trapped within the membrane-coated filter. When the pressure drop across the filter becomes too high for proper operation (or alternatively on run number basis), the outlet valve closes and air is purged through the filter while the heating elements raise the coupon temperature high enough to oxidize the soot within the filter. After a predetermined period of time, the air purge is discontinued and power to the heating elements turned off or down. The system is then ready to run a new test.

This approach has two technical advantages over the present approach. First, the smaller size of the proposed filter coupon will improve system response. With less volume between the source and analyzer, the lag time between exhaust gas generation and analysis is decreased. Second, if the filter is regenerated before each test, the amount of particulate on its surface will be nil. Any

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[4] After presentation to the SERDP Technical Thrust Area Working Group in May 2000, SERDP staff requested that CeraMem not pursue development of this application unless strong links to DoD needs could be established. CeraMem has stopped any development of this application using SERDP funding.

condensable gas adsorption on the remaining particulate is then very small and uniform from run to run. This will improve the analytical performance of the overall system.

Figure 11. Particulate-laden gas analyzer product concept (not to scale)
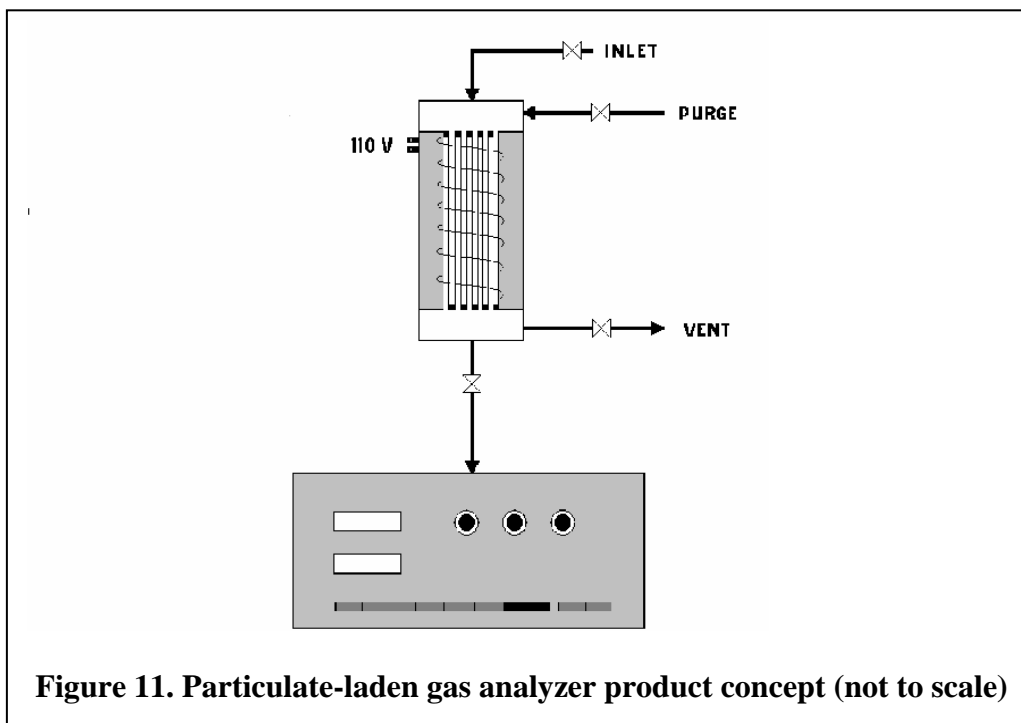
The economic feasibility analysis for the regenerable filter concept is shown in Table 3. Based on the estimated filter assembly price, annual O&M costs, and five-year life, the annual cost of the proposed concept to the end-user is 3 to 12 times less expensive than the present system. This clearly shows the potential benefit of the approach.

Table 3. Regenerable Filter Economic Feasibility Analysis

Item	Comments	Cost (to end user), $
Filter assembly & controls	1" x 2" filter	3,360
Yearly Power Consumption	250 days, 4 runs per day	28
Yearly Maintenance	3% of Capital	100
Annualized Filter System Cost	5 year life	800

5.0   Summary

The prices of three different filtration products that are being developed under the present program were estimated. The estimated prices are sensitive to a number of factors but most sensitive to monolith material and catalyst coating costs. Hot gas filtration systems incorporating

these filters have prices typical of filtration systems on the market based on price in $/ft^2. This is because most of the cost is usually in the system hardware.

Three potential applications were analyzed for preliminary economic feasibility. The use of passively regenerated gas filters for treating exhaust gas from jet engine test cells was proposed using a novel gas extraction system and a separate filtration system. The annualized costs of removing particulate matter and VOCs indicated that the process was probably not economic. The use of the new filters was compared to standard diesel trap technology for a portable generator set application. If the new filters are proven to have enhanced capabilities, these attributes may compensate for the small to modest price increase for aftertreatment systems incorporating them. Lastly, a new application for regenerable, hot gas filters was evaluated. It was found that the use of small filter coupons with in-line regeneration could substantially reduce the cost of prefiltration prior to engine exhaust gas analysis using advanced analytical instrumentation. In general, the technology to be developed may be economically feasible for a number of DoD applications but the technology needs to be developed and field tested to determine real-world utility.

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Oak Ridge Filter Test Facility

Date: **17 January, 2001**

To: **CeraMem Corp.**
                Mr. Robert Goldsmith   goldsmith@ceramem.com

CC: **Mr. Karsten Nielsen, Mr. Duane Adamson**

From: **Dave Crosby**

Pages: **1 of 2**

Subject: **ORFTF Tests**

Test Report

On January 9th 2001 the Q-76 Aerosol Penetrometer was powered up, calibrated and operated IAW procedure ATI-FTF-404 Rev.0. The test methodology used is IAW Mil-STD-282.

This penetrometer was modified by installing a regenerative high-pressure blower between the test chuck and the flowmeter sections. This blower was required to add the boost necessary to overcome the unusually high resistance of the filters to be tested. A special test adapter plate was machined to accommodate the new filter assembly. The filters supplied were tested at both the 100% of rated flow (125 cfm) and at 20% of rated flow (25 cfm) for both aerosol penetration and airflow resistance.
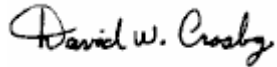
We first tested the complete assembled unit provided by CeraMem, which contained filter element # 14. We then tested the other unit, which was assembled in Owings Mills, MD under the direction of Karsten Nielsen. This unit contained element # 2.

We then disassembled these units IAW Karsten's procedures and reassembled 4 other elements In the ASTM housings and tested them. The results of these tests are attached on our standard ORFTF Test form.

These tests were performed under Purchase Order # 27777 and were witnessed by Westinghouse Savannah River representative Duane Adamson. Duane also carried back with him an assembled unit containing filter element # 7 and also filter element # 13 for additional work at WSR Co.

If you should have any questions or require additional information, please don't hesitate to contact me.

Sincerely;

David W. Crosby
VP and ORFTF Project Manager

ATI – Air Techniques International

## OAK RIDGE FILTER TEST FACILITY
F I L T E R   T E S T   R E P O R T

	PAGE 1 OF 1

CUSTOMER CeraMem Corp.	TEST CRITERIA	NUMBER ORDERED 6	DATE RECEIVED 6
	DOP PENETRATION .03 % @ 20% .03% @ RATED FLOW .of rated flow		
PURCHASE ORDER NO. 27777	RESISTANCE Inches, W.G. @ 100 %	NUMBER RECEIVED 0	DATE TESTED 9 Jan 01
FILTER MODEL NUMBER Prototype	RATED FLOW N/A	NUMBER ACCEPTED 6	DATE SHIPPED
	SPECIFICATION DOE STD 3027-97		

MANUFACTURER CeraMem Corp	TEST CONDITIONS	REJECTS	
FILTER DESCIPTION Ceramic Prototype	TEMPERATURE 71 °F	TEST FLOW (ACFM) 125/25	PENETRATION	RESISTANCE
RATED FLOW (ACFM) 126.1	BAROMETRIC RESS. 29.47 IN., hg.	Humidity in % RH 21	DAMAGE	OTHER

ITEM No.	FILTER SERIAL NUMBER	INSPECTION RESULTS	TEST RESULTS		
			RESISTANCE Inches WC	@ 100% FLOW	@ 20% FLOW
1	14	125 cfm	15.0	0.15	
2	14	25 cfm	1.2	0.14	
3					
4	2	125 cfm	11.6	4.8	
5	2	25 cfm	2.4	4.8	
6					
7	13	125 cfm	18.0	.044	

8	**13**	**25 cfm**	**3.6**	**.039**	
9					
10	**10**	**125 cfm**	**15.4**	**0.13**	
11	**10**	**25 cfm**	**3.6**	**0.18**	
12					
13	**9**	**125 cfm**	**18.8**	**.077**	
14	**9**	**25 cfm**	**4.4**	**0.21**	
15					
16	**7**	125 cfm	**21.8**	**.037**	
17	**7**		**4.8**	**.073**	
18					
DISTRIBUTION			TESTED BY: *David W. Crosby*		

Filter Housing

1/9/2001 13:12
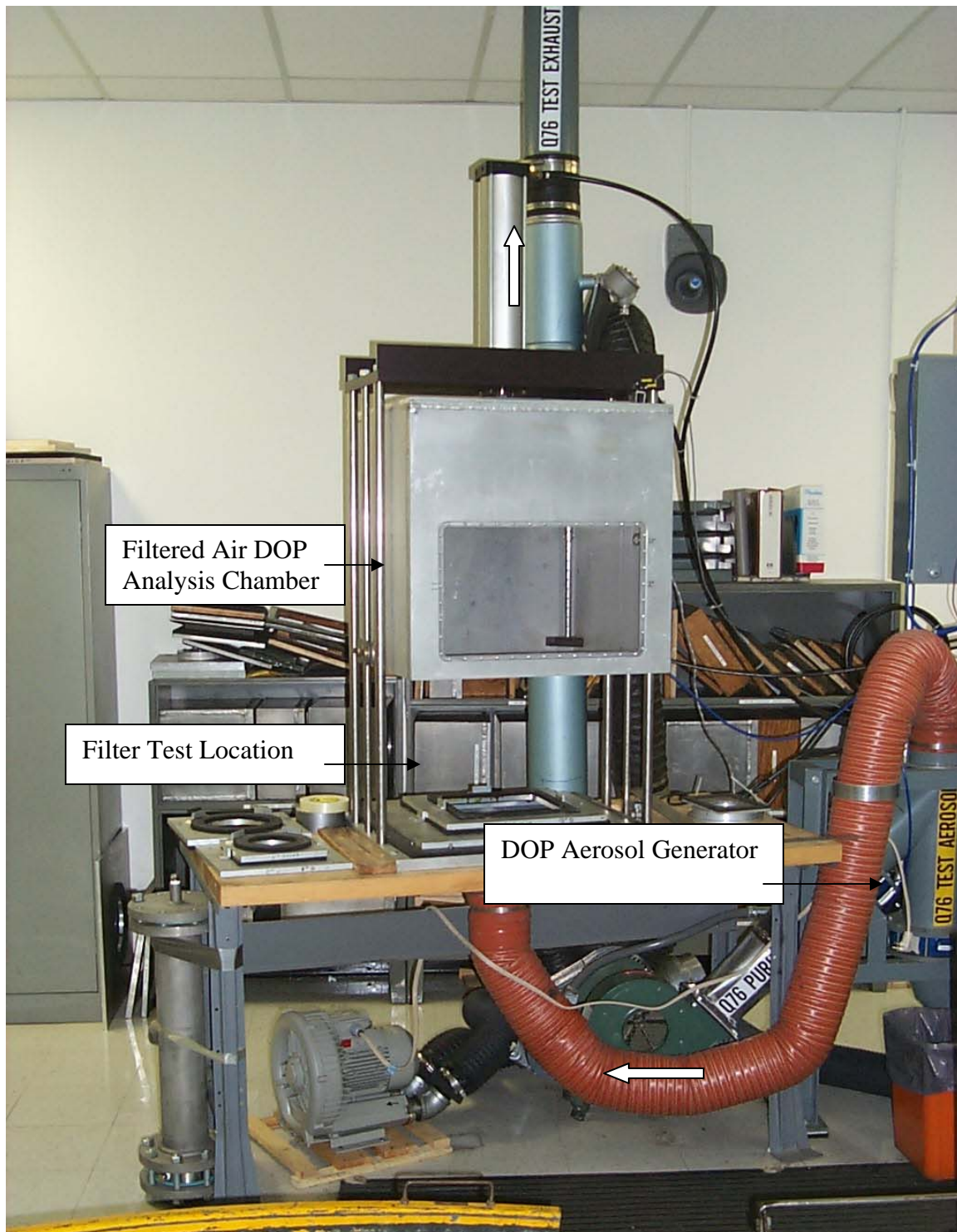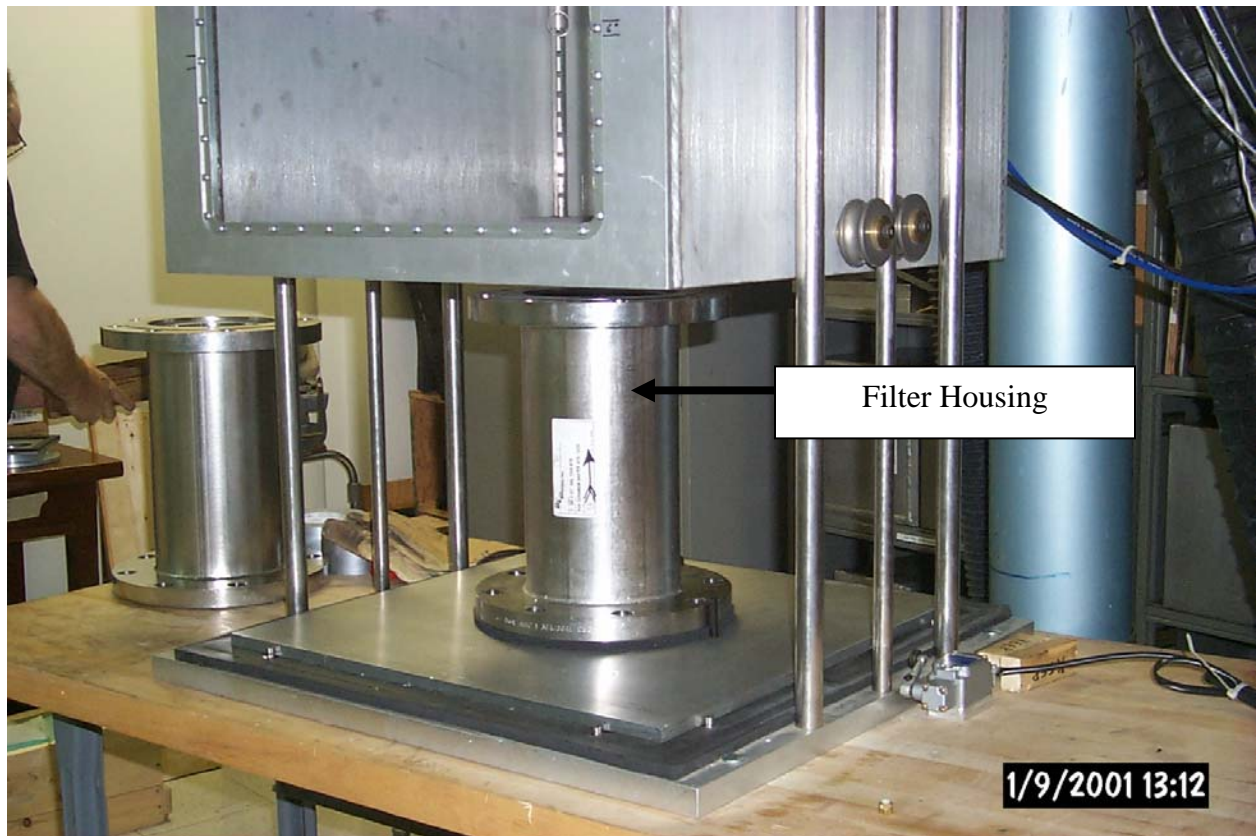
APPENDIX C. TEST RESULTS FROM UND EERC

October 9, 2000

Mr. Bruce Bishop
Vice President
CeraMem Corporation
12 Clematis Avenue
Waltham, MA 02154

Dear Mr. Bishop:

Subject: Final Report on the Evaluation of CeraMem Filters
　　　　 CeraMem Purchase Order No. 2137

　　　　 CeraMem has indicated it will not test all of the filters planned in the original proposal. Therefore, this will serve as the draft final report on the project to evaluate CeraMem filters.

　　　　 A ceramic CeraMem filter (designated SC-131) was evaluated at the EERC, and a technical progress report was submitted to CeraMem in November of 1999 detailing the results. The report included the filter's pressure drop characteristics, fractional particulate collection efficiency, and $PM_{2.5}$ (particulate matter less than 2.5 microns in diameter) mass emissions under a given mass loading. A second filter (designated SC-254) was evaluated in May 2000, and this report presents the results from the evaluation of that filter.

　　　　 A schematic of the bench-scale system used to evaluate the filter was presented in the previous report. The nominal filter area was 0.4 ft^2, and the target face velocity was 10 ft/min. To achieve this face velocity required a total gas flow of 240 ft^3/hr, which was maintained with three mass flow controllers and checked with a Ritter wet test meter. All flow rates were adjusted prior to injection of particulate. The differential pressure across the filter was measured with a 0–50-in. W.C. magnehelic pressure gage with pressure ports upstream and downstream of the filter. A Tri-Jet aerosol generator manufactured by TSI Inc. was used to generate a NaCl aerosol, with dry nitrogen used as the carrier gas. Near real-time inlet and outlet particle sizing measurements were made using a Model 33 aerodynamic particle sizer (APS 33) and a Model 3934 scanning mobility particle sizer (SMPS), both manufactured by TSI, Inc.

　　　　 Figure 1 is a plot of the pressure drop across the filter as a function of time. Over the 4-hr period of the test, the pressure drop increased from an initial value of 19.75 in. W.C. to a high of 24.5 in. W.C. The initial pressure drop was higher than expected, but this may be due to the pressure taps being located roughly 3 diameters upstream and downstream of the filter.

　　　　 Regardless of the initial value, the pressure drop increased by 4.75 in. W.C. over the 4-hr test period, which is similar to the 4.4-in. W.C. increase observed with the SC-131 filter.

Modified U.S. Environmental Protection Agency (EPA) Method 5 samples were taken at the inlet and outlet of the filter to determine total mass loadings. The inlet sample was conducted after the 4-hr test period, and the outlet sample was collected for the duration of the test. The inlet mass loading (based on the modified Method 5 sample) was 16.3174 mg/m^3. However, the mass loading predicted by the combined APS–SMPS data was 76 mg/m^3. The Method 5 inlet mass loading is lower than anticipated by the APS–SMPS data and the calibration of the Tri-Jet aerosol generator. The inlet mass loading for the SC-131 filter was also low, but it was suspected this was because of nonisokinetic sampling. It is not clear why the modified EPA inlet mass loading is low. The inlet and outlet APS–SMPS sampling was isokinetic. The outlet mass loading was 0.0267 mg/m^3, which is almost an order of magnitude lower than the mass loading of 0.1242 mg/m^3 reported for the SC-131 filter. For the outlet sample, the entire gas stream passed through the Method 5 filter, and represents the mass loading over the entire test period. A corresponding outlet mass loading based on the APS–SMPS data cannot be calculated because the outlet mass loading changed over time. On the basis of the APS–SMPS inlet and outlet data, the overall mass collection efficiency at the end of the test was 99.99%, which is significantly better than the 99.77% collection efficiency of the SC-131 filter. On the basis of the EPA Method 5 data, the overall collection efficiency for the SC-254 filter was 99.84%, which is significantly better than the 97.86% collection efficiency for the SC-131 filter.

Figure 2 is a plot of the PM$_{2.5}$ emissions as a function of time for the filter. The PM$_{2.5}$ emissions were fairly constant for this filter, averaging 0.0085 mg/m^3. For comparison, the SC-131 filter started at 0.185 mg/m^3 and decreased to 0.001 mg/m^3 at the end of the test.

Figure 3 plots the particulate collection efficiency as a function of particle diameter (fractional collection efficiency) for the filter at the beginning of the test and at the end of the test. The collection efficiency for the filter was high from the beginning of the test and showed no sign of increasing with the development of a residual dust cake. The data for the smallest particles may not provide a good representation of the collection efficiency because they are at the low end of the instrument's range. If the collection efficiency for the smallest particles is not counted, then the most penetrating particle size is roughly 0.03 μm, with a collection efficiency of roughly 99.965%.
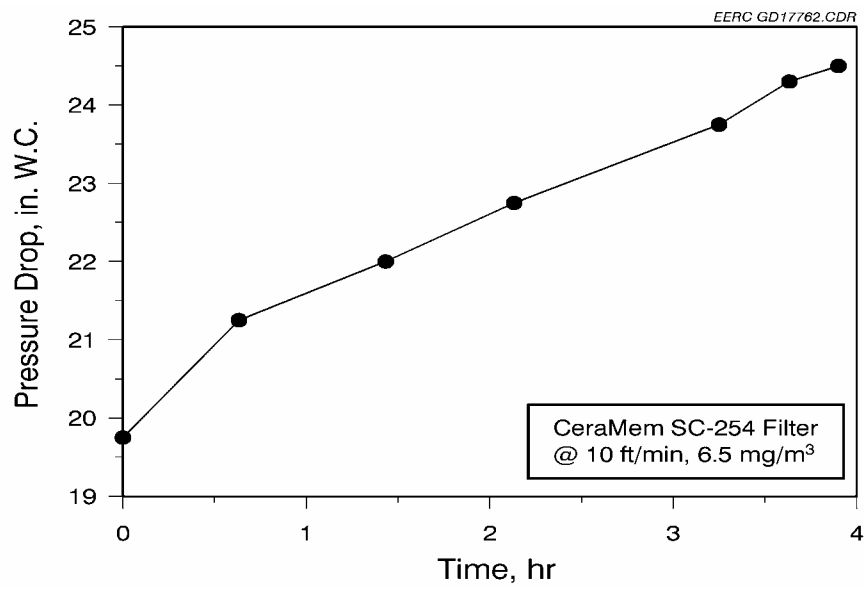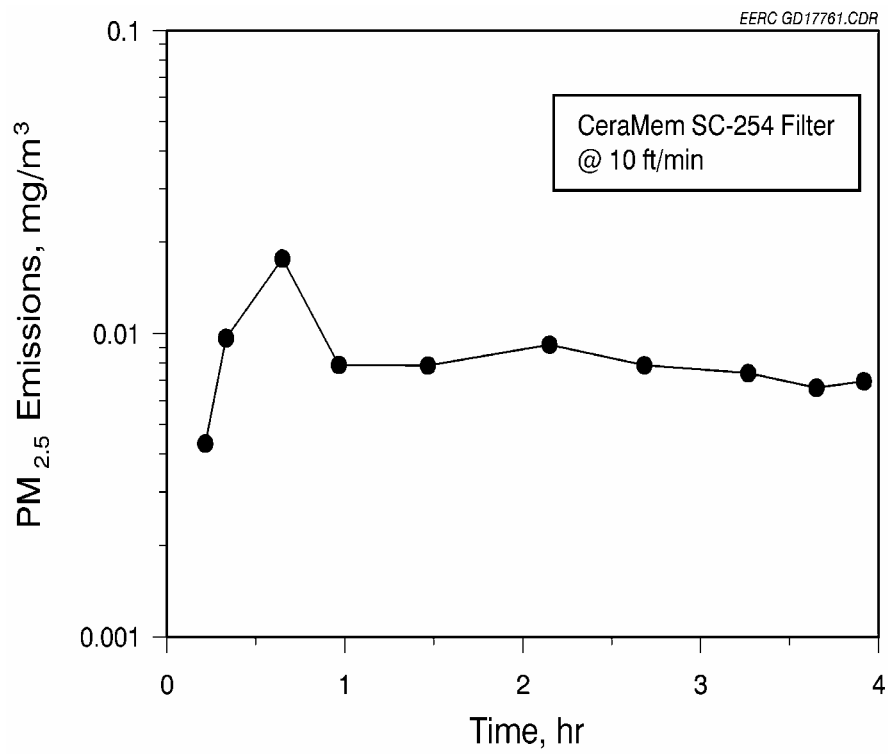
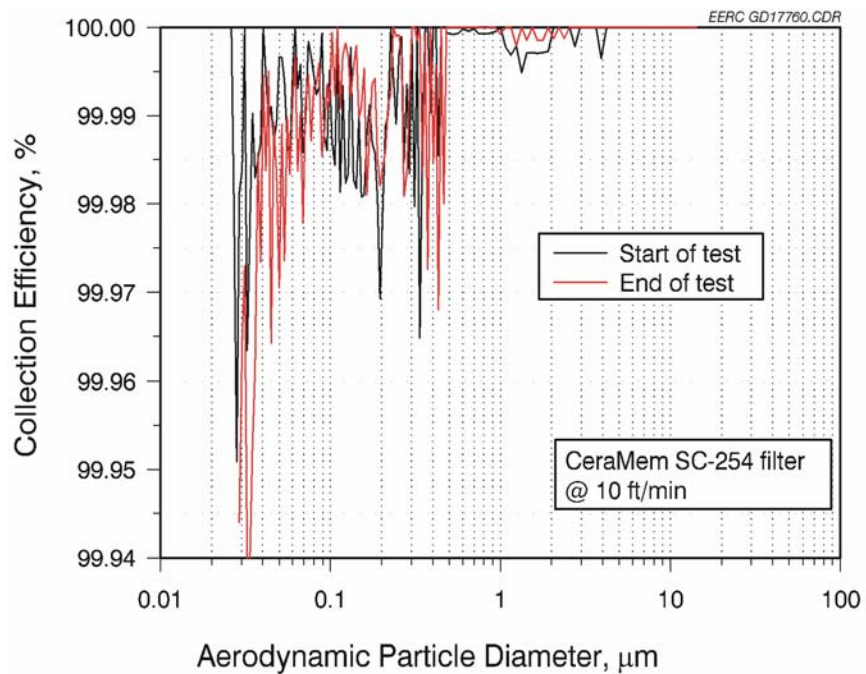**Figure 1**

Figure 2

Figure 3

If you have any questions or require further information, please contact me at (701) 777-5034 or gdunham@undeerc.org.

Sincerely,

Grant E. Dunham
Research Engineer

GED/kmd

APPENDIX D

STATEMENT OF WORK

Membrane Coated Filter Characterization with Fuel Borne Catalysts

Submitted to
CeraMem Corporation
12 Clematis Avenue
Waltham, MA 02453

Prepared by
André Boehman and Richard Steinberger
The Energy Institute
University Park, PA 16802

Scope of Work to be Performed

The activities to be performed under this project involve characterization of the regeneration, pressure drop and filtration efficiency of membrane- and non membrane-coated SiC diesel particulate filters prepared by CeraMem Corporation. The work is to be performed under CeraMem's existing contract with the federal SERDP Program. The Energy Institute's involvement with the study consists of two phases of study, with an optional third phase. The Project Plan consists of four tasks that involve: preparation of facilities: examination of membrane coated filter (MCF) and uncoated filter (UCF) operation with low sulfur fuel: examination of MCF and UCF operation with a fuel-borne catalyst and (optional) examination of asymmetric membrane coated filter (AMCF).

Statement of Work

The project consists of four primary tasks.

Task 0 Preparation of Engine Stand for Membrane Coated Filter Characterization (8 Weeks)

Under this task we will add pressure sensing, additional temperature sensors and analytical strategies for particulate measurement for characterization of the diesel particulate filters on an engine stand. We will also assist CeraMem with engineering an installation of the ECS "Combifilter" system for regeneration.

Subtasks:
0.1   Complete PID (define PID) control of engine speed
0.2   Complete installation of exhaust system pressure sensing
0.3   Develop methodologies for using the R&P 1105 TEOM particulate mass analyzer on the VW TDI engine in addition to the existing BG-1.
0.4   Develop/select analytical methodology to measure sulfate content in the particulate matter
0.5   Perform soot emission mapping of VW TDI engine to verify particulate emission versus load correlation, as well as, other criteria pollutant emissions

0.6 Configure exhaust system for either "quick connect" addition/removal or bypass of the "Combifilter" system to permit warm-up of the engine with the filter off-line – a two-way valve supplied by ECS.

0.7 Connect the filter and regeneration system thermocouples to the Micromax data system for real-time recording of system temperatures. You should be aware that installation of the internal thermocouples will depend on whether we get the Combifilter electrical regeneration option.

Task 1 Filter Pre-Conditioning and System Conditioning (2 weeks)

A non-membrane coated SiC filter and a membrane coated SiC filter will be loaded with 8g/L (~19g) of soot and then regenerated, with this process repeated at least three times, to stabilize the filters. The engine will be operated at 1800 rpm and 25% load. Exhaust temperatures and system pressures will be monitored. Another important measurement will be the time required to fill the filter to the 19 g loading limit and comparison of this value with the particulate emission correlation.

Subtasks:
1.1 Stabilize the uncoated SiC filter (1 week)
1.2 Stabilize the membrane coated SiC filter (1 week)

Task 2 Filter Performance Using Electrical Regeneration (6 weeks)

Under this task we will perform measurements of filter loading and regeneration at 1800 rpm and 25, 50, 75 and 100% load, regenerating the filter between each setting and repeating the cycle 3 times on both the non-membrane coated and membrane coated filters. During these tests, we will monitor exhaust backpressure, filter pressure drop, fuel consumption, and gaseous and particulate emissions upstream and downstream of the filters and downstream of the oxidation catalyst in the ECS Combifilter system. In addition, particulate composition upstream and downstream of the MCF/UCF will be examined with the R&P 5100 (a DTA device) to assess particulate composition. To assess particulate size distribution (mass of PM2.5 or greater), either SEM analysis of lightly loaded particulate filters or impactor sampling will be employed. During this "snap test", particulate mass (via the TEOM instrument, VOCs (via hot-FID) and CO (via FTIR) will be monitored to assess the amount of volatile release during the transient.

Subtasks:
2.1 Examine uncoated filter performance over a sequence of loads regenerating between modes (1.5 weeks)
2.2 Perform snap test to look for volatile release after partial loading of the uncoated filter (1.5 weeks)
2.3 Examine membrane coated filter performance over a sequence of loads regenerating between modes (1.5 weeks)
2.4 Perform snap test to look for volatile release after partial loading of the membrane coated filter (ditto above)

Task 3 Membrane and Non-Membrane Coated Filter Characterization With Fuel Borne Catalyst (5 weeks)

Under this task, the primary tests will be to verify the Break Even Temperature (as indicated by the catalyst supplier) for effective regeneration using a fuel borne catalyst selected by CeraMem. Break Even Temperature (BET) will be assessed by cooling from an elevated exhaust temperature (via engine load reduction) until the filter pressure begins to increase continuously. The BET will show the operating exhaust temperature where the filters can achieve continuous regeneration. During the BET tests, multiple cycles of temperature ramps will be examined, but only limited emissions measurements for determination of air fuel ratio in the exhaust will be monitored. These BET tests will provide information on the impact of filter configuration and thermal properties on regeneration. Engine out emissions will be examined to assess the impact of the fuel borne catalyst in the engine combustion process. Internal thermocouple sensors will provide information on the manner of regeneration, the burn front passing through the trap and whether sufficient material has accumulated in the trap to facilitate a regenerative burn.

Subtasks:
3.1   Determine BET with sufficient repetition of the measurement to assess experimental uncertainty for un-coated SiC filter using the fuel borne catalyst, 50 ppm sulfur fuel and at 1800 rpm engine speed (1.5 weeks)
3.2   Determine BET with sufficient repetition of the measurement to assess experimental uncertainty for membrane coated SiC filter using the fuel borne catalyst, 50 ppm sulfur fuel and at 1800 rpm engine speed (1.5 weeks)
3.2   Determine the impact of filter membrane on internal filter temperatures during induced catalytic regeneration (2 weeks) – both filters for comparison

Task 4 Impact of Fuel Sulfur on Catalyst Performance (2 weeks)

The impact of fuel sulfur level on the effectiveness of the fuel borne catalyst will be considered as an experimental variable. All initial tests will be performed with a low sulfur fuel, provided to Penn State by Mobil Oil Company, containing roughly 50 ppm sulfur. To this baseline fuel, sulfur will be added to the fuel via the addition of di-tert-butyl disulfide (e.g., 4vol% addition of this compound provides 300 ppm equivalent sulfur weight to the fuel), to achieve up to a maximum of 20,000 ppm sulfur content in the fuel. BET testing will be performed at 150 ppm (CARB) and 300 ppm (nominal US domestic) sulfur content, and then in increments (1000, 3000, etc) up to 20,000 ppm or until catalytic activity ceases. The number of incremental steps will be determined during testing based on the impact of sulfur on the BET. Likely, little response to sulfur content will be observed until an elevated sulfur content is achieved (e.g., 100 ppm). As BET rises due to catalytic inhibition by the sulfur, a point may be reached where soot oxidation cannot be achieved at the exhaust temperatures the engine produces at 1800 rpm. Above that sulfur level, only the 20,000 ppm case will be examined. During these fuel sulfur variations, the trap will be operated at a somewhat elevated temperature (e.g., above 400°C) and as fuel sulfur is increased the steady state pressure drop may increase.

Where no detectable effect on pressure drop is observed, then a BET determination will be performed.

Subtasks:

4.1 Examine the impact of fuel sulfur steady state pressure drop and, if necessary, on BET for the CeraMem MCF in increments of fuel sulfur level starting at 150 and 300 ppm and ending at 20,000 ppm (2 weeks)

Task 5 Characterization of an Asymmetrically Membrane Coated Filter (3 weeks) (*optional*)

Under this task, for an asymmetrically plugged membrane coated SiC filter (AMCF), we will determine the regenerability, particulate collection efficiency, operational pressure drop and required fuel borne catalyst loading for effective regeneration using a fuel borne catalyst selected by CeraMem. This will be accomplished by repeating Tasks 2 and 3 as appropriate.

Subtasks:

5.1 Examine AMCF performance over a sequence of loads regenerating between modes (1 weeks)

5.2 Perform snap test to look for volatile release after partial loading of the AMCF (1 weeks)

5.3 Determine BET with sufficient repetition of the measurement to assess experimental uncertainty for the AMCF using the fuel borne catalyst, 50 ppm sulfur fuel and at 1800 rpm engine speed (1 weeks)

Task 6 Travel to CeraMem for Reporting and Presentation of Results

Under this task, as required by CeraMem and the Technical Advisory Committee for the research program, personnel from Penn State will travel to CeraMem and present results from the project. Performance of this task will be subject to the needs of CeraMem and the availability of the Penn State personnel

Subtasks:

6.1 Travel to Boston (CeraMem headquarters) or other location as needed for technical presentation of results

Reporting

Time to accomplish data analysis has been included in all tasks and subtasks, so that reports will be prepared and submitted in Microsoft Word and all data will be compiled in Microsoft Excel spreadsheets. Monthly summary progress reports will be submitted electronically to CeraMem and its staff. A final report will be submitted in printed form and via a CD-ROM.

Budget

The project budget is $76,271 for a period of performance from February 1, 2001 to August 31, 2001. Testing should be complete by August 10, 2001. Without Task 5, which is optional depending on availability of suitable materials, the project end date can move forward three weeks.

The budget breakdown is as follows:

	Total
Personnel and Fringe	29,852
Materials & Supplies	7,500
Travel	2,850
Services	5,250
Equipment	12,001
Indirect	18,817
TOTAL	76,271

The budget breakdown by task is:

	Total	Task 0	Task 1	Task 2	Task 3	Task 4	Task 5	Task 6
Personnel and Fringe	29,852	9,185	2,296	6,889	5,741	2,296	3,445	
Materials & Supplies	7,500	2,308	577	1,731	1,442	577	865	
Travel	2,850	-	-	-	-	-	-	2,850
Services	5,250	1,615	404	1,212	1,010	404	606	
Equipment	12,001	12,001	0	0	0	0	0	
Indirect	18,817	5,427	1,357	4,070	3,392	1,357	2,035	1,180
TOTAL	76,271	30,536	4,634	13,902	11,585	4,634	6,951	4,030

Payment Schedule

The following shows a timeline for completion of the tasks and the billing/invoicing schedule.

	Task 0	Task 1	Task 2	Task 3	Task 4	Task 5	Task 6
Time from Start of Contract (weeks)	8	10	16	21	23	26	TBD
Invoice Amount	30,536	4,634	13,902	11,585	4,634	6,951	4,030

APPENDIX E: PRELIMINARY TEST PLAN OUTLINE FOR INCINERATOR TRIALS

PROGRAM OBJECTIVES

The first objective of the incinerator flue gas treatment program is to demonstrate that a CeraMem membrane-coated filter (MCF) has a superior filtration efficiency thus providing a means of achieving the newly proposed PM2.5 regulation for DoD applications. Higher filtration efficiency addresses concerns regarding the release of μm-sized heavy metal particulates. This increase in filtration efficiency must be accomplished without an unacceptable pressure drop across the flue gas filter system. This includes the need to demonstrate that MCFs can be repeatedly regenerated without long-term degeneration of pressure drop performance (i.e., progressive and permanent clogging of membrane pores with fine ash). A second objective includes the reduction of dioxin concentrations in the flue gas. A third objective is the reduction of volatile organic compounds (VOCs) and CO by means of a suitable catalyst coating on the filter substrate.

BACKGROUND

Burning of paper and plastic waste, both on Navy vessels and for land-based applications, provides an acceptable means of achieving a 90 percent volume reduction of these wastes. Based on an NRC study for the US Navy, the emissions of primary concern are dioxins and heavy metals. Concentrations of VOCs and CO may take on a greater importance for land-based applications – particularly near large military personnel or civilian population concentrations. These latter emissions can, however, be effectively controlled by more precise control of the combustion process.

Reduction of heavy metals can be achieved by higher filtration efficiencies – particularly for the small particulate size fraction. CeraMem's membrane coated filters (MCF) are particularly well suited for this latter task. MCFs made of SiC can be safely used at flue gas temperatures well above the recommended temperatures (~1000°C) for the types of incinerators being considered here.

There are two mechanisms for the formation of dioxins. The first is a homogeneous mechanism in which the products of incomplete combustion are converted to dioxins at temperatures in the range of operation of the incinerators (~ 1000°C). The second is a heterogeneous mechanism in which the products of incomplete combustion react on the surface of the fly ash in the temperature range of 250° to 350°C. While dioxin production can be reduced by more complete oxidation of its precursors, the more effective method involves the reduction of particle loading and residence time in the temperature range in which dioxins are produced. This is achieved by rapidly quenching the flue gas to temperatures below 250°C and/or removing the particulates at temperatures above 350°C.

Dioxins are quite resistant to oxidation, which why they persist so long in the environment. Pure dioxins require temperature in excess of 800°C for their destruction. There is also evidence to suggest that when dioxins are bound to particulates they can survive temperature

above 1100°C. In the case of the MCF, the VOC catalyst would be applied to the membrane support (i.e., filter substrate) and is located downstream of the filter membrane. The catalyst would, therefore, have no impact on dioxins deposited in the filter cake. Since the homogeneous production of dioxins takes place at temperatures above 800°C any reduction of the precursors of dioxin by the VOC catalyst will have no impact on dioxin production. That is, only the heterogeneous reaction takes place at temperatures below 350°C and this latter reaction will be effected most by removal of the (catalytic) ash by filtration. It is questionably whether the VOC catalyst will have any effect on gas phase dioxin passing through the filter since temperatures > 800°C are generally required for thermal destruction of this compound.

OVERVIEW OF TEST PROGRAM

The development of the MCF is well advanced and is not described further here (Bob's job). The filter system, however, requires the construction and testing of a venturi-based back-pulse regeneration system as shown schematically in Figure 1. This system will be tested and its performance optimized at CeraMem before installation in the pilot plant filtration unit.
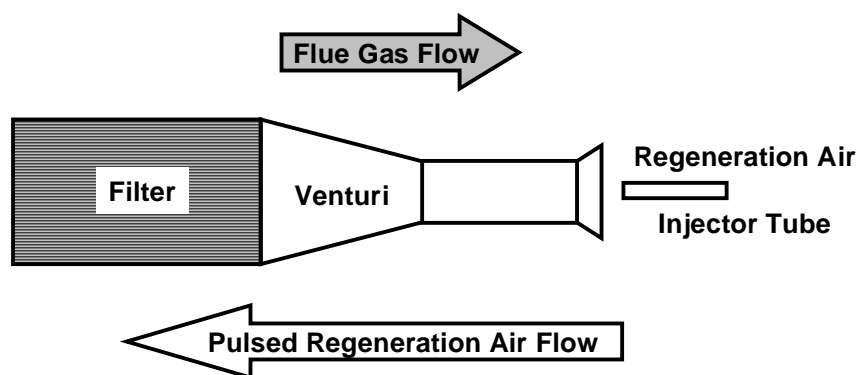

Figure 2: Schematic of Back-Pulsed Venturi Regeneration System for MCF. Arrows indicate direction of flue gas flow through the MCF during normal operation and the direction of pulsed air flow during offline regeneration. The filter assembly is mounted in a vertical orientation in the filter housing so that the regeneration air blows the filtered flue ash downwards into a collection hopper.

The down-fired combustor (DFC) at Penn State will be used to simulate an incinerator. Using coal as fuel, the DFC is capable of flame temperatures up to 1500°C and has a modular heat exchanger that can reduce flue gas temperatures to 100°C (see Figure 2). CeraMem has a pilot plant capable of housing up to three 5.66" diameter x 8" long MCFs at temperatures up to about 370°C. A slip stream from the DFC will connected to the pilot plant on order to simulate an incinerator flue gas filtration system. The fly ash collection filters will be regenerated off line using a pulsed back-flow technique. The MCFs will be may coated with a suitable catalyst to study the oxidation of VOCs, CO and other emissions such dioxins.

The DFC is primarily coal fired but is sufficiently versatile that other combustible materials such as paper and ground plastic could be added to the combustion stream to simulate a range of incinerator feed materials. A key element in this study will be the ability to produce particulates that are representative in size distribution and composition to those produced in a waste incinerator. This is an essential requirement in order to demonstrate the ability of the filters to efficiently remove μm-sized particulates. Upstream injection of VOCs, dioxins and other materials can also be used to more accurately control flue gas composition upstream of the catalyzed MCF system. This is particularly important for studies involving dioxins where trace analysis of flue gas is a very time consuming, expensive and not always reliable in terms of accurate analytical results.
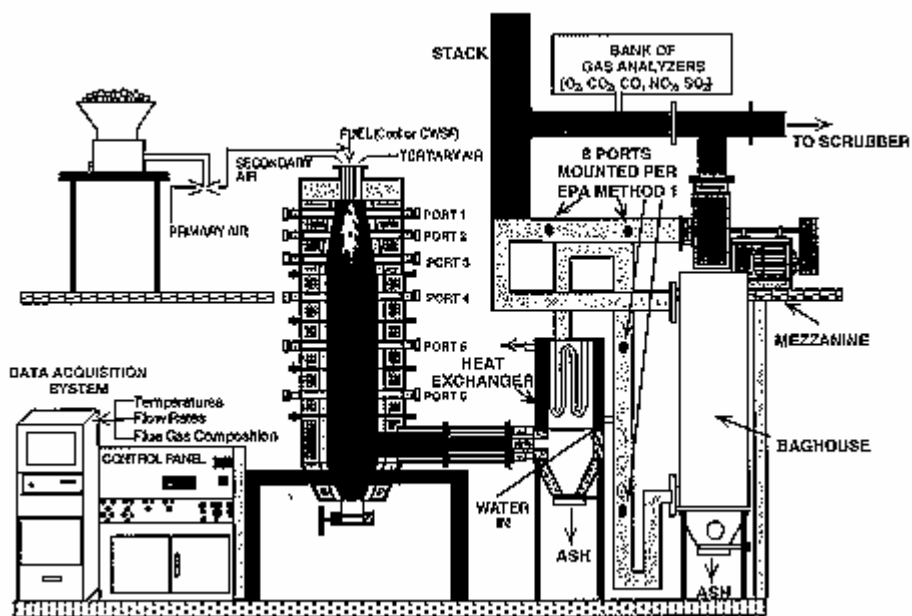

Figure 2: Schematic Diagram of the 500,000 Btu/h Down-Fired Combustor

The test program will have three phases which cover three different contaminant groups. Phase 1 will address particulate trapping efficiency – particularly for small μm sized particles - and MCF pressure drop characteristics and pressure drop recovery after filter regeneration. Phase 2 will address the efficiency of the catalyzed MCF for oxidizing VOCs and CO. Phase 3 will address dioxin production and/or destruction.

SCOPE OF WORK

Phase 1: The back-pulsed venturi MCF regeneration system, shown in Figure 1, will be constructed, tested and optimized at CeraMem prior to installation in the filtration unit at Penn State.

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Particles in the flue gas from an incinerator are typically in the range of 1 to 50 μm in size and have a composition similar to the ash residue. In addition, < 1μm sized particles enriched in toxic metals such as lead, cadmium and arsenic are formed by the vaporization of these metals in the combustion chamber and their condensation in the cooler region downstream. Ideally filtration could take place at high temperature, however, pressure drop considerations favor filtration at lower temperatures (and gas velocities). To achieve reduction of both toxic metals and dioxin production, however, filtration should take place at temperatures above 350°C. If the filter is coated with a VOC catalyst then temperatures about 350°C will enhance oxidation reactions.

The fuel feed and/or combustion air feed rates for the DFC will be adjusted to provide an outlet temperature of about 1000°C. The heat exchanger cooling rate will be adjusted to provide an inlet temperature to the filters of about 350°C. The filters will be regenerated offline at the end of each operating period or as pressure drop across the MCFs dictates. The following parameters will be measured on a continuous basis:

- heat exchange inlet and outlet temperature;
- filter unit inlet and outlet temperature; and
- filter unit inlet pressure and pressure drop across the filters.

Periodic measurements of the following parameters will be made:

- filter inlet and outlet particulate concentrations and size distribution;
- ash composition; and
- flue gas composition (e.g., CO, SO_4, VOCs as required).

Particular attention will be taken to accurately determining the pressure drop across the filters after each regeneration in order to ascertain the efficiency of the MCF regeneration process and equipment.

Phase 2: This phase of the program addresses the impact of the MCF on reducing dioxin production and/or its release to the environment. Filtration removes particulates that catalyze dioxin production (heterogeneous reaction). Filtration also removes particulates that have dioxin bound to them and/or the filter cake may act as a condensation site for dioxin (i.e., removal of dioxin produced by the high temperature homogeneous reaction). The effectiveness of this latter mechanism can unambiguously be assessed by injecting a known concentration (i.e., larger than that normally present) of dioxin upstream of the filter. Gas samples will be collected using the AmbStack sampling train. These samples will then be analyzed using the CALUX bioassay kit.[5] Differences between upstream and downstream dioxin concentrations should reflect the impact of the filter and filter cake for removing dioxins produced by the high temperature homogeneous reaction. This is a control or bench mark experiment.

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[5] An AmbStack gas sampling train is used to concentrate the dioxin on a Polyurethane foam. The dioxin is then extracted from the foam and analyzed using CALUX bioassay method.

The impact of filtration on heterogeneous dioxin production will be investigated by injecting of a suitable organic waste simulate containing chlorine as part of the fuel to the DFC in order to enhance dioxin production. In the first test, the filter will be operated at 350°C to remove the heterogeneous reaction phase catalyst (i.e., the particulates). In a second test the filter will be operated at 100°C to remove the products of both reactions. Again, up- and down-stream sampling of the flue gas using the AmbStack sampling train will provide samples for analysis using the CALUX bioassay kit. By comparing the concentrations upstream of the filter for the two experiments it will be possible to ascertain the production rate of the two mechanisms. The downstream concentrations will determine the efficiency of removal of the dioxin from the gas stream and the effectiveness of the filter for minimizing the heterogeneous reaction.

Phase 3 (optional): This phase of the program will be directed at determining the effectiveness of the catalyst-coated MCFs for oxidizing VOCs and CO. It is likely that there is sufficient CO in the flue gas to measure the effectiveness of the catalyst without further CO injection. Further CO injection into the flue gas upstream of the MCFs could, however, be used if warranted. The effectiveness of VOC oxidation will be measured by injection of C_7H_8 (600 – 700 ppm) into the flue gas upstream of the filter. These tests will be carried out over a sufficient time period to assess the long-term durability of the catalyst in this type of environment. If injection of dioxin upstream of the filter in Phase 2 indicates that the filter is not 100% effective in removing dioxin, then a challenge of dioxin could be used to test the effectiveness of the VOC catalyst to oxidize any dioxin passing through the filter. Up- and down-stream sampling of the flue gas using the AmbStack sampling train will provide samples for analysis using the CALUX bioassay kit.

REPORTING OF DATA

All lab data will be recorded in Microsoft Excel spreadsheets to facilitate post experimental analysis. Written reports will be submitted using Microsoft Word. These documents will be transmitted by e-mail to the project manager. Progress reports will be submitted with data on a month-end basis.