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Ceramic Filters for Removal of Particulates From Hot Gas Streams

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DISCLAIMER

Ceramic Filters For Removal Of Particulates From Hot Gas Streams

CONTRACT INFORMATION

Contract Number DE-FG02-90ER80896

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Period of Performance May 20, 1991 to May 19, 1993

Schedule and Milestones

FY 93 Program Schedule

	S	O	N	D	J	F	M	A	M	J
Ceramic Filter Development										
Scaleup To Full Size Filters						- COMPLETED -				
Conduct Durability Tests						- COMPLETED -				
Conduct Regeneration Test						- COMPLETED -				
Field Demonstration										
Select Test Site	—	—								
Design/Build Hardware		—	—							
Conduct Field Tests				—	—	—	—	—	—	—
Full Size Filter Design/Costing							—	—	—	—
Final Report									—	—

OBJECTIVES

The primary goal of this Phase II SBIR program is to demonstrate the performance of a new ceramic filter in removing particulate matter from hot gas streams produced in advanced coal conversion processes. The specific objectives are threefold:

(1) Development of full size ceramic filters suitable for hot gas filtration;

(2) Demonstration of ceramic filters in long term (ca. 1000 hrs) field trials; and

(3) Development of full-scale hot gas filter system designs and costs.

BACKGROUND INFORMATION

This paper describes a novel ceramic filter being developed under a U.S. DOE SBIR Phase II grant for hot gas filtration. The need for hot gas cleanup in advanced coal conversion processes is well documented and extensive development is being undertaken to develop and demonstrate suitable filtration technologies. In general, process conditions are (a) oxidizing or reducing atmospheres, (b) temperatures to 2000°F, and pressures to 20 atm. The most developed technology entails the use of candle filters, which suffer from fragility, temperature limitations, and high cost. The filter under development in this program offers the potential to eliminate these limitations.

PROJECT DESCRIPTION

Ceramic Gas Filter Description

The construction of the ceramic filter is based on the use of porous honeycomb ceramic monoliths. These high surface area, low cost materials are widely used as catalyst supports for automotive catalytic converters. The monoliths have a multiplicity of "cells" (passageways) which extend from an inlet end face to an opposing outlet end face. The cell structure can be round, square, or triangular, and the cell "densities" can vary from 25 to 1400 cells per square inch. Porosity of the honeycomb material can be from below 30% to over 50%. The mean pore size can range from about 4 to 50 microns. The superior properties of commercially available honeycomb ceramic monolith materials make them ideally suited for applications requiring high thermal stability, mechanical strength, and corrosion resistance. These rigid ceramics have been used for years as automotive catalyst supports where conditions of high vibration and thermal cycling are encountered in a combustion gas environment. Other applications of these materials as catalyst supports include emission control systems such as catalytic incineration and NO_x SCR.

The monolith structure used for catalyst support material is readily adapted to function as a filter to remove particulate matter from diesel engine exhaust. Unlike the catalytic convertor application in which automotive exhaust flows in a crossflow mode through the honeycomb cells, the diesel particulate filter (DPF) operates as a dead-ended filter. The carbonaceous soot in the exhaust gas is filtered on and within the cell walls of the monolith. This is achieved by modifying the monolith structure by plugging every other cell at the upstream face of the device (Figure 1) with a high-temperature inorganic cement. Cells which are open at the upstream face of the monolith are plugged at the downstream face. Exhaust gas is thereby constrained to flow through the porous cell walls, and at appropriate intervals, the filter is cleaned by burning off the entrapped soot.

A variety of monolith sizes is available for DPF devices. Typical monolith characteristics are a square cell shape, a cell density of 100 cells/square inch, a cell wall side of 0.083", and a cell wall thickness of 0.017". DPF devices operate at least in part as depth filters. The pore size of the cell wall material is quite large (20-35 microns), and fine particulates enter and plug the cell wall structure. This leads to pore plugging by particulates and makes regeneration by backpulsing difficult, if not impossible.

CeraMem has developed technology for applying thin ceramic membrane coatings to honeycomb monoliths to produce crossflow microfiltration (MF) and ultrafiltration (UF) membrane modules. These membrane coatings have pore sizes substantially smaller than the pore size of the monolith support material, and the membrane coatings function as surface filters. Pore sizes of the different CeraMem membrane coatings are in the range from 1.5 microns (the coarsest) to 40 Angstroms. The coating technique involves filling the cells of the monolith with a liquid medium ("slip") containing a mixture of ceramic powders, dispersants, and polymeric binders. The pore structure of the monolith absorbs water from the slip, forming a cake of particles on the walls (surfaces) of the passageways. After a defined absorption time, excess slip is drained from the passageways, and the coated monolith is dried and fired (up to 2400°F) to bond the ceramic membrane particles to themselves and the cell wall surfaces. Membrane pore size and porosity are determined primarily by the particle size of the ceramic particles used in the slip.

Ceramic Filter For Hot Gas Particulate Removal

To filter particulates from hot gas, CeraMem modifies the DPF device described above by applying a ceramic MF membrane to at least the inlet cell wall surfaces. This creates a composite filter which can be operated as a backpulsable surface filter. The thin, membrane coating has a pore size approximately 100-fold finer than that of the monolith support (Figure 2). Thus, the retention efficiency of the filter for fine particles is determined by the membrane pore size. By keeping the membrane coating thin (ca. 50 microns) the resistance to gas flow (pressure drop) is kept acceptably low. Yet, it is possible to use a large-pored, low-resistance support for the membrane. Because it is coated by the membrane, the pore size of the support does not affect particle retention and the pore structure of the support does not become plugged by particulate matter.

In operation, ash-laden gas flows into the membrane-coated inlet cells. Particulates are collected in the inlet cells and the filtered gas exits the module via the downstream cells. As particulate material accumulates, pressure drop increases to a preselected level at which time the filter is cleaned by online backpulsing from the downstream end of the filter.

RESULTS

Properties Of Filters Developed In First Year Of Program

During the first year of the program ceramic filters with the sizes shown in Table 1 were developed. The compactness of the filter relative to other gas filters is evident from the data of Table 2. This compactness leads to very compact systems as the filters can be installed in a closely packed array in a filter vessel. Both the filters and total filter systems are expected to have costs much lower than those of traditional hot gas particulate filtration systems.

Figure 3 shows a photograph of the largest of the filters produced to date. Figure 4 shows a photograph of two filters sealed in a housing assembly by means of wrapping the filters in a ceramic fiber mat and fitting the filters into a steel housing. The compressed mat serves to cushion the filters and hold them firmly in place as well as to provide a particle-tight seal. An assembly of 16 filters in 4 steel housings is shown in Figure 5. Four assemblies of backpulse venturis are shown nestled on top of the filter housings. This array of filters contains approximately 600 ft^2 of filter area in a cube about 2 ft on a side. The assembly would be mounted directly into a tube sheet in a hot gas filter vessel.

Table 1. Characteristics Of Ceramic Gas Filters

Dimensions Of Ceramic Gas Filter	Filter Area, Ft^2
4.66" Diameter x 5" Long	5
5.66" Diameter x 6" Long	10
5.9" Side (Square) x 12" Long	37

Table 2. Comparison Of Ceramic Filter With Other Filters

Filter Type	Filter Dimensions	Filter Area (ft^2)	Area/Volume Ratio (ft^2/ft^3)
CeraMem	5.9" x 5.9" x 12"	37	155
Fabric Bag	6" Dia. x 20' Long	31	8
Candle	6cm Dia. x 1.5m Long	2.8	19
Crossflow	12" x 12" x 4"	8.3	25

The retention efficiency of the ceramic filter is greatly increased by the addition of the membrane coating, as can be seen in the data of Table 3. The retention of filters, with and without the membrane coating, has been measured for dilute aqueous suspensions of narrowly-sized alumina particles. The turbidities of a feed suspension and the initial filtrate (collected before a filter cake can build) are measured.

Table 3. Retention Data For Filters With And Without Membrane Coating

(Filtration Tests With Aqueous Alumina Suspensions)

Sample	Alumina Particle Size, Microns	Retention, %
Uncoated	5	17
Membrane-Coated	5	99+
	3	99+
	0.5	95

The uncoated monolith passes particles with a size of 5 microns almost completely. In contrast, the membrane-coated filter has substantially complete retention for 5 micron particles and very high retention for 0.5 micron particles. Given the additional particle capture mechanisms in gas filtration, a membrane-coated filter with these liquid retention properties can be expected to have substantially complete retention for submicron particles.

Figure 6 shows flow/pressure drop data for 12" long filters with and without the membrane coating. The membrane coating increases the resistance over that of an uncoated filter about two- to three-fold. For the uncoated filter, the increase in pressure drop with increasing filtration rate is due to pressure drop for gasflow in the filter passageways. For membrane coated filters the primary resistance to gas flow is that of the membrane coating itself.

Thermal Durability Testing

Thermal cycling and shock tests have been undertaken at CeraMem to evaluate the thermal durability of the filters. A 4.66" diameter x 5" long filter was fabricated and tested for air flow/pressure drop and alumina retention characteristics prior to exposure to thermal cycling and shock.

The filter was placed in an electric kiln and thermal cycling tests performed in which the kiln was fired at the maximum firing rate to heat up as quickly as possible to 1650°F. The kiln was held at 1650°F for one hour and kiln power was shut off, allowing the kiln to cool to ambient temperature. Each heating cycle took about 45 minutes and the total cycle required about 12 hours. After 35 cycles the filter was removed and inspected. No change was observed in its visual properties.

The filter was then subjected to thermal shock testing in the electric kiln. Pulses of cold compressed air were discharged onto the face of the filter through a stainless steel tube connected to the compressed air source. A timer/solenoid valve assembly was used to expose the filter to 1 second pulses of cold air at 4 minute intervals. During the interval between pulses, the filter inlet face temperature recovered to the kiln temperature of 1650°F after experiencing a drop of several hundred degrees during the cold pulse. The filter was subjected to 1005 pulses and removed from the kiln for evaluation. No visual effects from the pulsing were observed. The thermal cycling and shock described above did not have any effect on filter performance as measured by air flow/pressure drop and retention for 0.5 and 5 micron alumina in the alumina suspension filtration tests described above.

Short-Term Field Test Results

To date, field tests of the ceramic filter for particulate removal have been conducted at seven sites on a variety of gas streams and under a variety of test conditions. Results of two hot gas tests will be presented below. In general, the following performance characteristics have been observed:

1. Filtration face velocity (equivalent to an "air to cloth ratio") for flue gas tests is comparable to that for pulse jet bags operating at the same pressure drop. In hot gas tests, flow-pressure drop characteristics have been observed to be comparable to those for other ceramic filters.

2. Complete regeneration by a simple backpulse technique is achieved; i.e., no increase in clean filter resistance over repetitive cycles is observed.

3. No plugging of the filter passageways by badly caking particulates is observed.

4. Essentially complete particulate removal, including submicron particulate matter, is achieved.

Tests At EERC/University Of North Dakota

In Phase I of this SBIR Grant, feasibility tests were conducted at the Energy And Environmental Research Center (EERC) at The University of North Dakota. The reactor system used for testing the filter was EERC's 100 lb/hr gasifier. As configured and tested, the reactor product gas was passed through a primary cyclone prior to introduction into a hot gas cleanup test loop containing a ceramic filter. A heated backpulse system was installed to provide hot backpulse nitrogen. Depending on the discharge pressure and duration of the backpulse cycle, the temperature of the backpulse gas entering the test loop decreased with time. Backpulse frequency and duration were controlled manually to achieve the desired number of pulses and pulse duration.

The fluidized bed calciner was operated in a hydrogen production mode using dolomite as the bed material and Wyodak coal as fuel. Steam and a small amount of oxygen were heated to approximately 1300°F and fed into the bottom of the reactor bed. During the runs, the differential pressure across the filter was monitored continuously and test system operators initiated filter backpulsing when the pressure drop across the filter began to rise at a fairly rapid rate. This occurred at a pressure differential reading of about 35" H_2O. Based on the gas flow, ash loading, expected ash bulk density, and filtration cycle time, the passageways of the filter were substantially filled with ash at the time of backpulse regeneration. During the tests, backpulsing was controlled manually, and typically 2 to 4 pulses of 1/2 second duration were used. The backpulsing nitrogen pressure was 75 psig at the nitrogen cylinders, and less (but not measured) at the ceramic filter.

Test conditions are summarized in Table 4 and representative pressure drop data over 9 cycles are shown in Figure 7. Within-cycle pressure fluctuations are associated with gasifier pressure variation due to a variable coal feed rate. The clean filter baseline differential pressure was maintained through these nine cycles, as well as over the entire test period.

Table 4. Test Conditions For Tests At University Of North Dakota

Filter Dimensions:	4.66" dia. x 5" long
Filter Area:	5 ft^2
Coal Type:	Wyodak
Face Velocity:	9 to 10 ft/min
Test Temperature:	1200°F (approx.)
Test Pressure:	2 atm abs.
Ash Content:	0.8 to 1 grains/DSCF
Typical Filtration Cycle:	50 minutes (2500 ACF)
Backpulse Conditions:	Offline
	Heated Nitrogen
	Four 0.5 second pulses

While feed ash loadings were determined accurately, attempts to measure the filtered gas ash loading were unsuccessful due to the design of the piping arrangement of the test loop, which resulted in ash introduction into the sampling loop during backpulsing. However, it is believed that the ash retention of the filter was substantially complete based on visual observations of the downstream side of the filter and piping system, which were completely clean.

A summary of the conclusions from the EERC tests is given in Table 5.

Table 5. Conclusions From Tests At University Of North Dakota

In 45 filtration/backpulse regeneration cycles in two week test period:

1. No change was observed in the clean filter pressure drop.

2. Ash removal efficiency appeared to be 100%.

3. No degradation was observed in any filter properties after repetitive thermal cycling.

Tests At Westinghouse Science And Technology Center

Short term feasibility tests were conducted at the hot gas test facility of the Westinghouse Science and Technology Center in Pittsburgh in December, 1991. In these tests Grimethorpe ash was injected into combustion gas and a series of filtration cycles was performed over a 4 day test period. The general test conditions are given in Table 6. A trace of filter pressure drop over a series of 10 cycles is shown in Figure 8. A comparison of filter pressure drop as a function of filter face velocity is given in Figure 9. Also shown is the expected pressure drop based on extrapolation from room temperature air tests at CeraMem, correcting for viscosity. These results were obtained with the filter before and after the Westinghouse test. The difference in filter pressure drop measured under simulated PFBC conditions from that extrapolated from ambient tests has not been resolved. Future tests will be conducted to reconcile this discrepancy.

Conclusions from the Westinghouse test are given in Table 7.

Table 6. Test Conditions For Tests At Westinghouse

Filter Dimensions:	4.66" dia. x 5" long
Filter Area:	5 ft^2
Fuel Type:	Gas
Face Velocity:	4 to 8 ft/min
Test Temperature:	1200°F (approx.)
Test Pressure:	100 psig
Ash Content:	3500 ppm Grimethorpe ash
Typical Filtration Cycle:	15 minutes
Backpulse Conditions:	Online Cold air @ 150 psig Three 0.5 sec. pulses

Table 7. Conclusions From Tests At Westinghouse

In 40 filtration/backpulse regeneration cycles in four day test period:

1. No filter plugging was observed.

2. Ash removal efficiency was about 100% (0.5 to 1 ppm ash in filtered gas).

3. Measured pressure drop was two-fold higher than expected, based on extrapolation from ambient air data. Reason not yet determined.

FUTURE WORK

Future Field Trials

An important objective of this program involves field demonstration of the filter in longer term tests. Unfortunately, the test site originally chosen for long term field trials in the Phase II program is not operational at this time. CeraMem is now actively seeking an alternative test site. A possible site is at METC treating hot gas from a 6" AFBC. Other possible sites are being evaluated, generally either ACFB's or PCFB's.

In addition to the DOE sponsored field tests, filters will be installed for pilot tests on several other hot gas streams, including:

- PFBC pilot plant in Europe, @1600-1800°F

- 600 ACFM flue gas from a medical waste incinerator, @ 1200-1800°F

- 160 ACFM hot gas from lead smelter, @ 1300°F

- 180 ACFM shale gasifier hot gas, @ 1000°F

- Sorbent injection/hot gas filtration for SO_2 removal, @1000°F

- Sorbent injection/hot gas filtration for H_2S removal, @1300-1400°F

Design And Costing Of Full Size Filter Systems

Over the next few months an Architect Engineer will be selected and subcontracted to perform a conceptual design study for the ceramic filter for hot gas filtration. Costs for conceptual designs will be compared with those for alternative systems using candle filters.

Schedule

Due to delay in selection of a field test site, it is anticipated that a no-cost program extension will be requested and that the program field tests will extend into the second half of 1993.

ACKNOWLEDGEMENTS

The development of this technology is being assisted by EPRI under Technology License Agreement RP1402-61. The tests at EERC were conducted under the direction of Mr. Jay Haley. The tests at Westinghouse were conducted under the direction of Dr. Thomas Lippert. Finally, the active assistance and support of Mr. Norman Holcombe of METC is greatly appreciated.

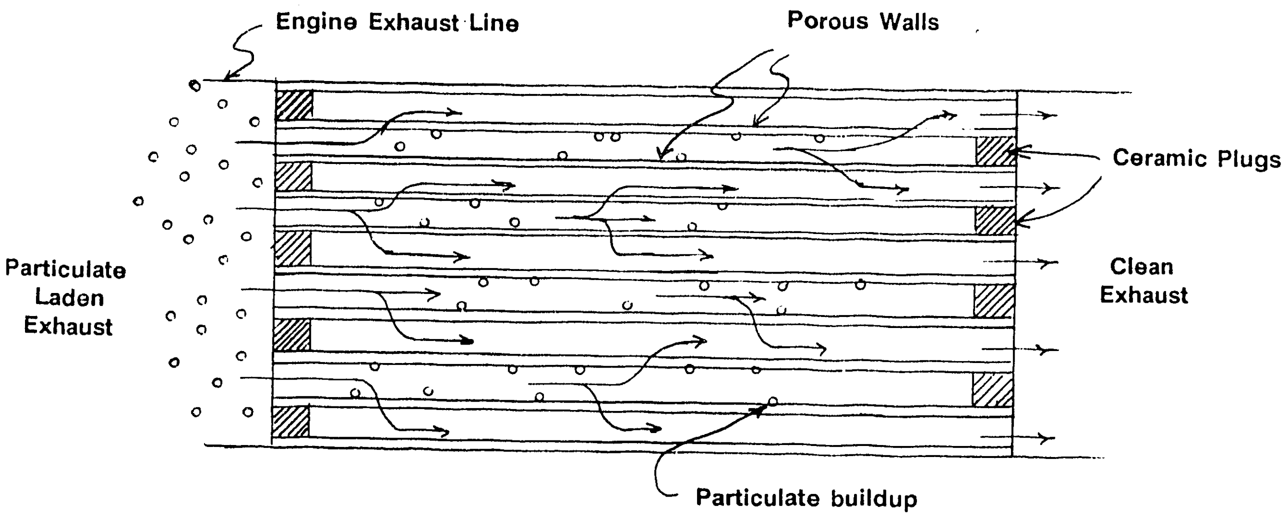

Figure 1. Honeycomb Ceramic Monolith Filter In "Dead-End" Flow Configuration

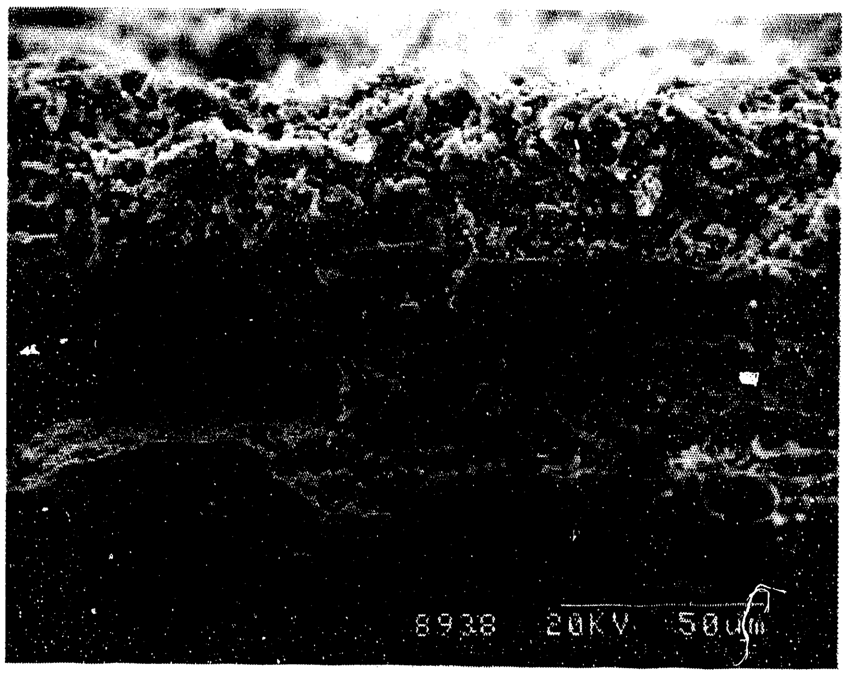

Figure 2. Scanning Electron Micrograph Of Membrane Coating On Monolith Support

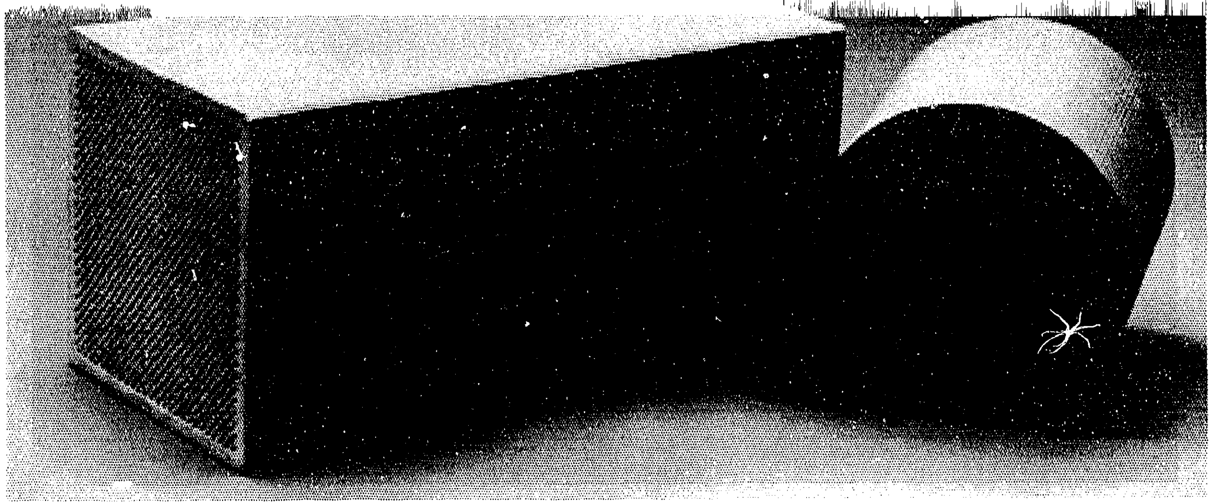

Figure 3. Photograph of Three Sizes of Ceramic Gas Filters

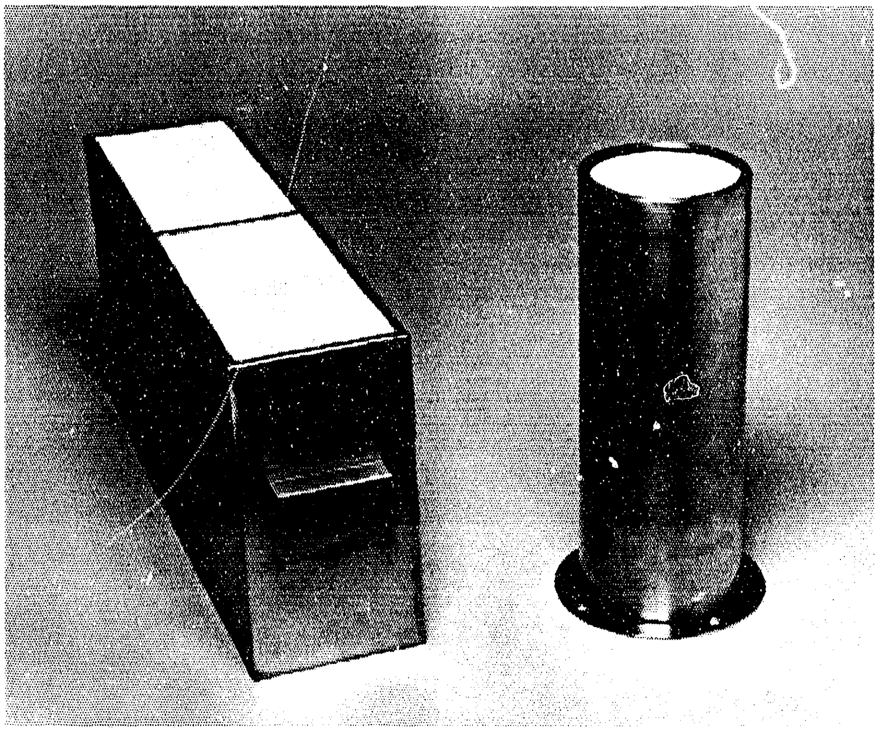

Figure 4. Photograph of Two Ceramic Filter Housings

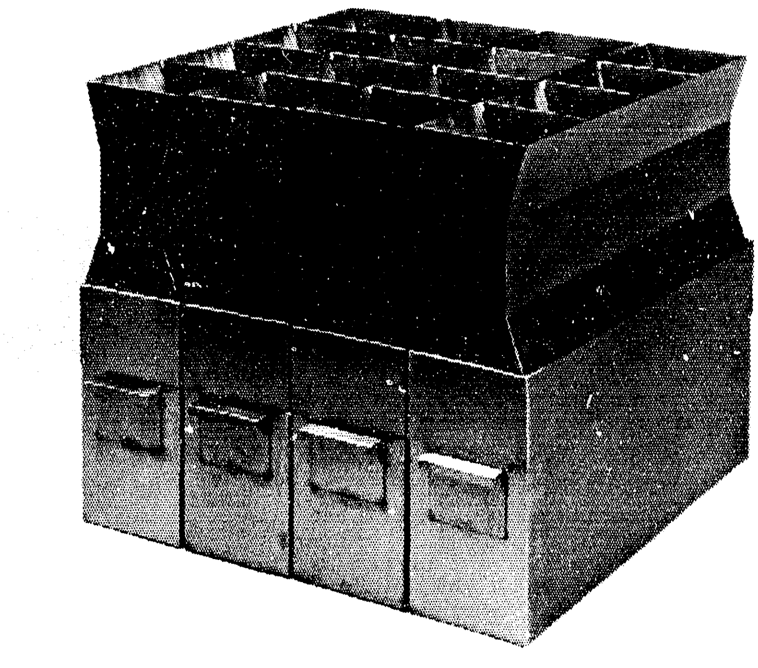

Figure 5. Photograph of Sixteen Ceramic Filter Assembly In Housings With Venturis

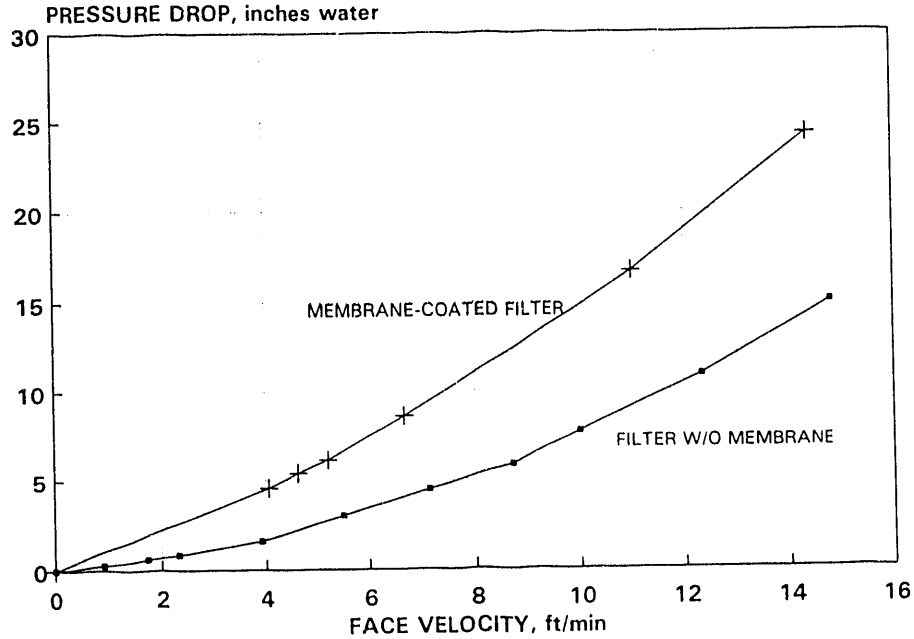

Figure 6. Pressure Drop Vs. Flow For Ceramic Filters With And Without Membrane
(Air @ 1 Atmosphere and 25 Degrees Centigrade)

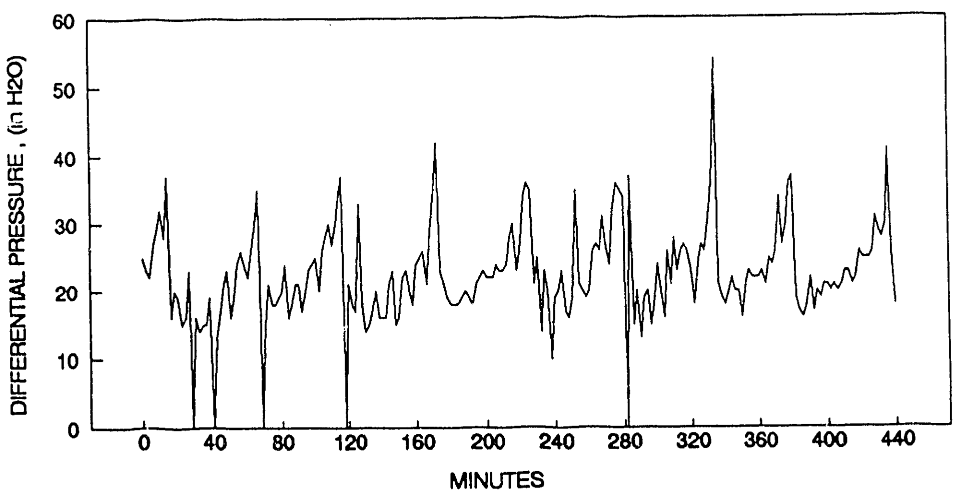

Figure 7. Pressure Drop Of Ceramic Filter In Tests At EERC (9 Cycles)
Typical Pressure Drop Across the Filter Element With Time

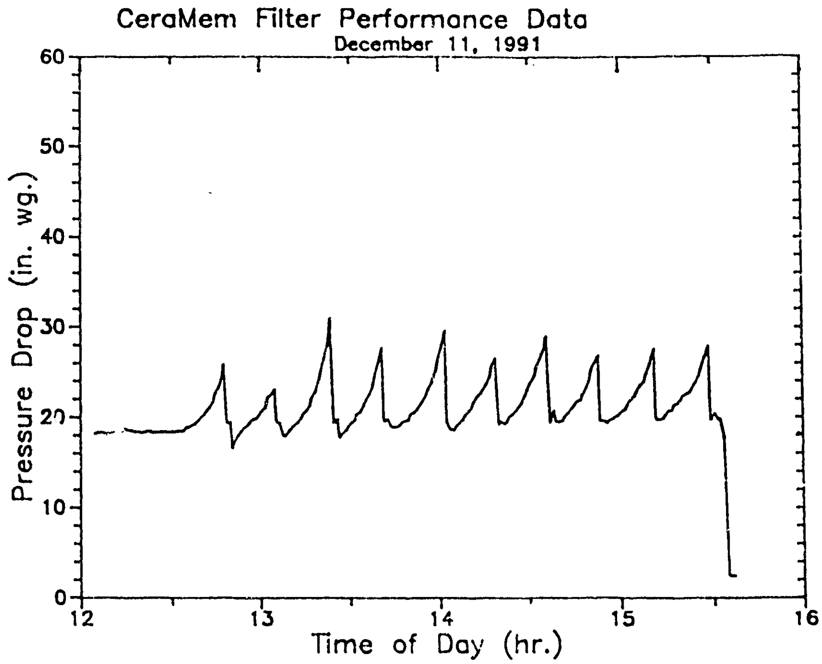

Figure 8. Pressure Drop Of Ceramic Filter In Tests At Westinghouse (10 Cycles)

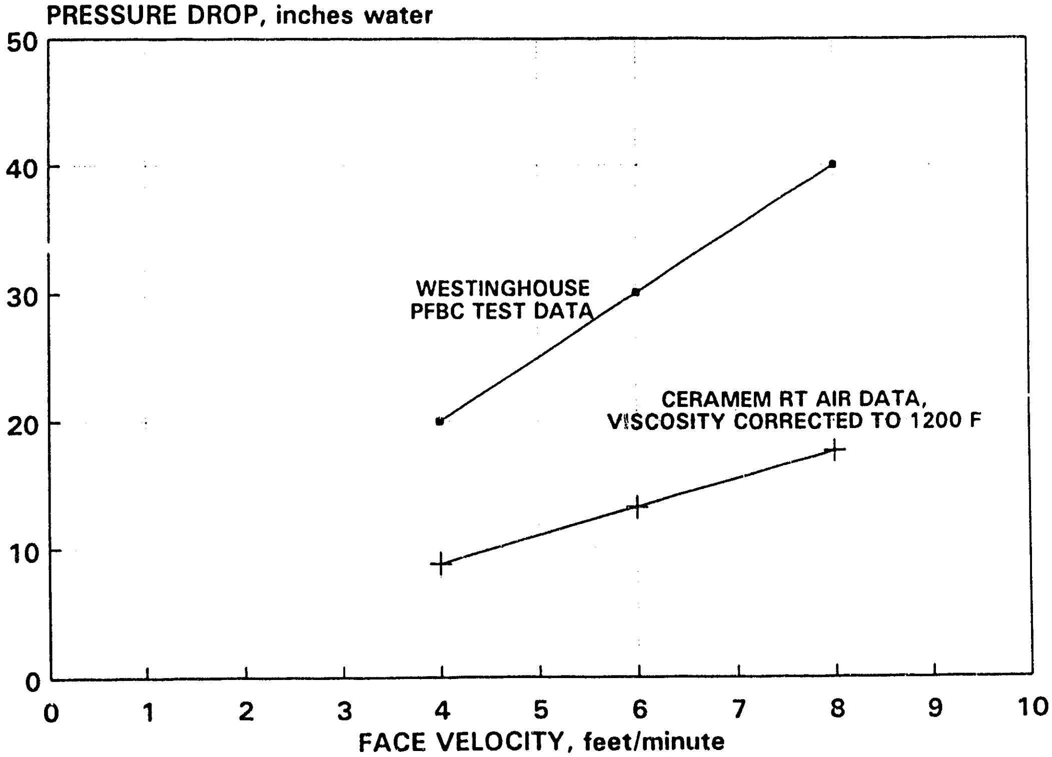

Figure 9. Pressure Drop Vs. Gas Flow In Tests At Westinghouse

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