CeraMem Filter Development Program

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Introduction

Advanced fossil energy power systems such as PFBC'S and IGCC's require high temperature barrier filters in order to operate effectively. In high temperature, high pressure combustors, ash must be removed from the combustion flue gas stream before it can enter a gas turbine. Ash particles will erode the turbine blades if the concentration and size of the particles is large enough, Even relatively small concentrations of particles can be very damaging, therefore, small numbers of seal or filter failures that allow bypass of the flue gas stream can not be tolerated for long periods of time. In addition, the filters must perform at very high temperatures or the power generation system will not operate at its optimum efficiency.

In gasification systems, the ash and char that is suspended in the syngas stream must be removed prior to the gas stream's introduction to other downstream air pollution control systems (e.g., hydrogen sulfide removal). The char, once captured, can be reinfected into the gasification reactors to increase the overall system efficiency. If the ash and char are not captured before the other downstream equipment, these solids can foul process piping or columns and cause downtime for the entire system.

The following paper describes, in general, a development program between CeraMem Separations and Foster Wheeler for a high temperature ceramic filtration system to be sold by their partnership, CeraFilter L.P. In addition, detailed test results of ceramic filters operating at conditions approaching those of high temperature combustors will be discussed. The successful development of this hot gas filtration system based upon the CeraMem ceramic-membrane-coated, monolithic filter will help address the critical challenges of particulate removal under the severe conditions imposed by advanced fossil energy power systems.

Research sponsored, in part, by the U.S. Department of Energy's Morgantown Energy Technology Center, under Grant Number DE-FG2-90ER80896 with CeraMem Corporation, 12 Clematis Avenue, Waltham, MA 02154; 617-899-0467

Objectives

To date, limited success has been achieved using various high temperature barrier filters under severe advanced combustion system conditions. Areas in which filters have had limited success include 1) mechanical and thermal stability of the filters, 2) high temperature sealing of the filters to the system, and 3) detrimental effects of system operation on the filters.

Mechanical and thermal stability of barrier filters has been a problem restricting the long term use of these filters at high temperatures. The present day leading technology, ceramic candle filters, has had a tendency to mechanically fail under PFBC conditions, These failures allow the bypass of flue gas and considerable amounts of damaging fly/ash particles. In addition, some candle materials have been susceptible to thermal shock and material degradation at high temperature.

Sealing of barrier filters into high temperature systems has been a challenge. A large number of seal designs have been developed to hold filters in place that are being exposed to relatively high differential pressures. Seals which fail allow particulate-laden flue gas to bypass the filter and thus lead to downstream system problems or failures,

The dynamics of filtration system operation are very complex. Two aspects that have been of concern are dust reentrainment and ash bridging. Reentrainment occurs if the ash pulsed from the filter is not allowed to settle into the ash hopper at the bottom of the filtration vessel and is reentrained back into the gas flow entering the filter. A considerable amount of cold modeling and vessel configuration testing has been performed to understand gas flows within filtration vessels in an effort to minimize fly/ash reentrainment. Bridging of ash between candle filters has been observed to occur in filtration systems operating for long periods of time. The bridging phenomena has caused mechanical failures of the candle filters.

The above issues have been considered in the preparation of the joint Product Development Program presently being undertaken by CeraFilter L.P. Therefore, the program is attempting to develop filtration systems with the following attributes:

- High particulate removal efficiency
- Refractory filter materials
- Resistance to oxidizing and reducing conditions
- Thermal shock resistance
- Mechanically rugged configuration
- Robust seal design
- Compact, modular design
- Optimum gas flow within the vessel to prevent ash retainment
- Minimum bridging potential between filters

Approach

As a solution to the above filter system problems, CeraMem has developed a compact, high temperature barrier filter that is resistant to catastrophic mechanical failure. This filter was developed under a US DOE Small Business Innovation Research grant (Grant Number DE-FG02-90ER80896) to clean hot flue gas from advanced coal conversion power processes.

The ceramic filter is based on a honeycomb ceramic monolith. This low cost ceramic material is widely used as a catalyst support for automotive catalytic converters. The monoliths are extruded in a variety of configurations and are commercially available from Coming, Inc. The monoliths have a multiplicity of "cells" or passageways which extend from an inlet end face to an opposing outlet end face. The cell structure is usually square and the cell "density" can vary from 4 to 16 cells per square centimeter,

The most commonly available monolith ceramic material is cordierite. The mean pore size of this material in the monoliths can range from about 4 to 50 microns with a porosity of 30% to 50%. The properties of these ceramic monoliths 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.

CeraMem modifies the honeycomb monoliths to produce high efficiency, dead-ended hot gas filters. First, a ceramic microfiltration membrane is cast onto the monolith passageway surfaces. The flow path through the monolith is then modified by plugging every other cell at the upstream face of the device with a high temperature inorganic cement. Cells which are open at the upstream face of the monolith are plugged at the downstream face. Gas is thereby constrained to flow from the inlet cells, through the porous ceramic membrane and cell walls, and then out the cells open at the downstream end. Any particles that flow into the filter with the gas stream are filtered out by the membrane. When the pressure drop across the filter rises to a predetermined level, compressed gas is pulsed back through the filter to dislodge the solids from the filter and they are collected in a hopper situated below the filter elements. A schematic of the filter is shown in Figure 1 and a photograph of an 18 cm diameter by 38 cm long filter is located in Figure 2.

This filter construction results in a number of advantages. First, the cordierite material results in a filter that is thermally stable in both oxidizing and reducing conditions, and is thermal shock resistant. The membrane coating produces a filter with high filtration efficiency, The monolith honeycomb structure, in conjunction with the housing which is similar to that of a catalytic converter, results in a mounted filter that is mechanically rugged and resistant to catastrophic failure. The honeycomb monolith also creates a filter with a large surface area to volume ratio which minimizes the size, and therefore the cost, of the filtration system. Consequently, the filters using this concept are capable of withstanding the severe environments of advanced power systems, have high filtration efficiency, and result in compact, cost efficient systems.

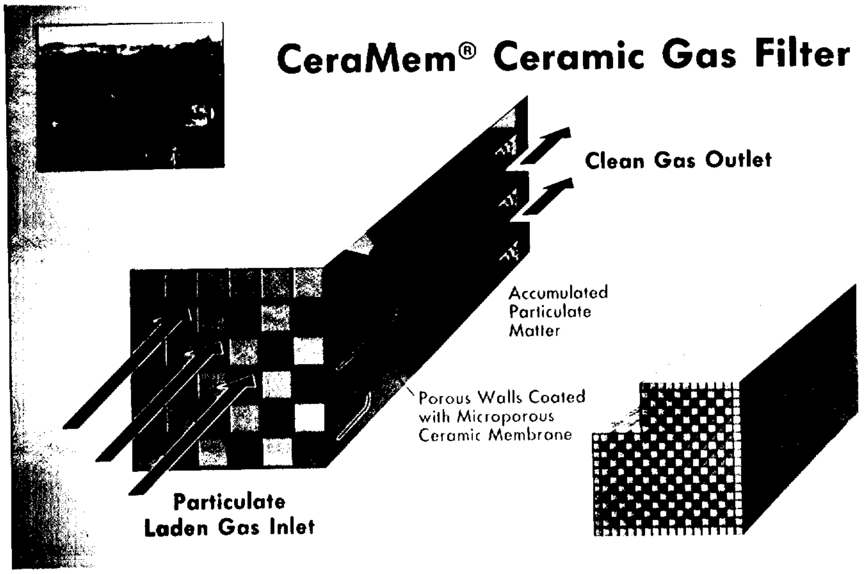

Figure 1. CeraMem® Gas Particulate Filter Schematic

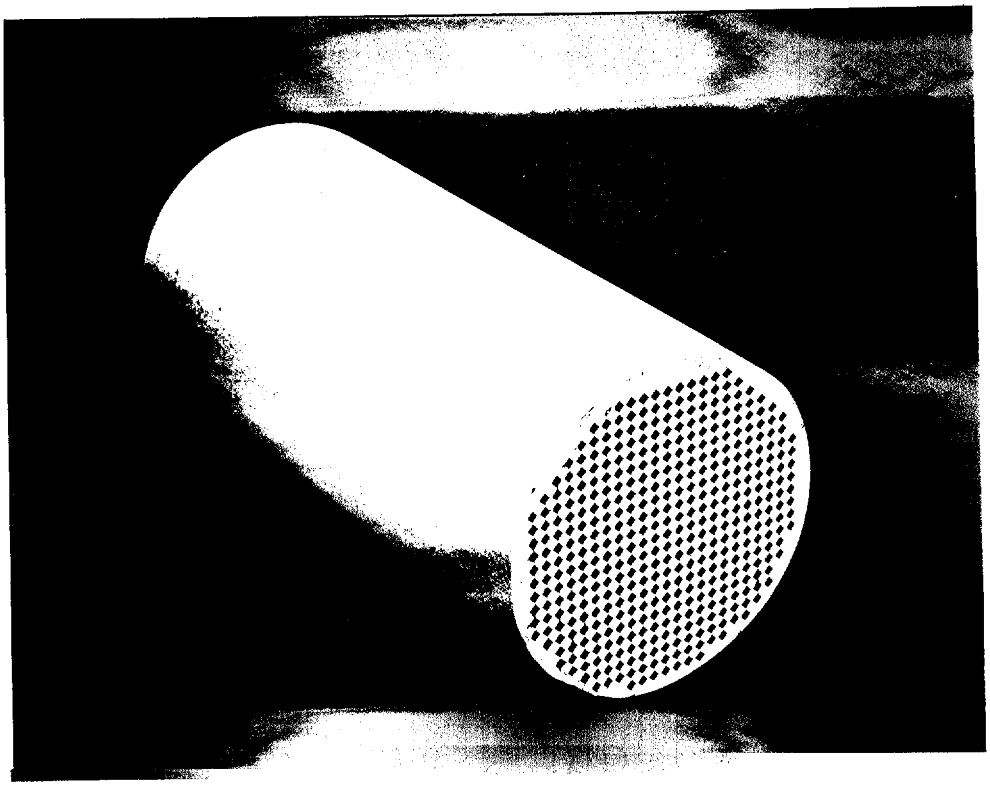

Figure 2. Photograph of 18 cm Diameter by 38 cm Long CeraMem® Gas Particulate Filter

CeraMem has entered into a partnership (CeraFilter L. P.) with Foster Wheeler to further develop the CeraMem filters and engineering systems for hot gas cleanup applications. In this partnership, CeraMem brings the filter technology while Foster Wheeler brings the system engineering capabilities and an indepth knowledge of the advanced power systems market. CeraFilter has put together a Product Development Plan through which CeraMem and Foster Wheeler have worked to bring high temperature filtration systems to commercialization.

Project Description

There are four sections of the Product Development Program. These are 1) Engineering Analysis, 2) Filter Development, 3) Filter Testing, and 4) System Design and Testing. There are a total of 16 tasks in the program, of which 14 have been completed. As part of the program, the intent is to conduct a pilot scale demonstration test. Upon successful completion of the demonstration test, the system will be ready for commercialization.

Engineering Analysis

In the engineering analysis section, several aspects of filtration systems were evaluated in paper studies. First, the conditions for design and testing were researched, The conditions selected for PFBC (non-topping) applications were a process temperature of 850 - 900°C, a process pressure of 12-17 atmospheres, and a dust loading of 50-180 g/m^3 while those for IGCC applications were 370- 540°C, 14-21 atmospheres, and 100-1500 g/m^3. It should be noted that the filtration temperature for gasification could increase depending on availability of materials, Second, the importance of backpulse gas consumption and filter pressure drop were investigated. It appears that these parameters will be more sensitive in PFBC applications than IGCC applications. Third, the effect of a pre-cyclone before a filtration system was evaluated. Fourth, the types of materials for various components throughout the system were selected. Using a variety of materials to meet the various operating conditions throughout filtration systems for both PFBC and IGCC applications is necessary in order to minimize material costs yet provide a reliable system.

Filter Development

The filter and filter mounting development tasks have been completed. First, the specifications and processes required to manufacture commercial scale filters (12” diameter x 15” length) have been developed. Prototype production runs have been completed at commercial yields to prove the manufacturing process. Second, the mountings used to connect the filter to the filtration system have been developed, The mountings have several subcomponents which have been evaluated to withstand the various operating conditions.

Filter Testing

Various aspects of the filters have been tested. First, a filter in its mounting has been tested successfully for its ability to hold differential pressure over many temperature cycles simulating actual operating conditions. Second, test scale filters have been evaluated for filtration performance during a 300 hour test on “live” flue gas at 870°C. This task will be described in detail in the results section. Third, filter performance was also tested in a cold model test in order to better understand the filtration process and formulate guidelines for successful gas filter regeneration. Fourth, a production scale filter was successfully tested in a simulated particulate-laden gas stream for 1,000 hours at temperature to determine if the mounted filter would regenerate over a long period of time. Last, a mounted filter was exposed to thermal cycling to determine if this configuration would be rugged enough to withstand actual filtration conditions. An examination following the tests indicated that no degradation of the filter or mounting construction occurred.

System Design and Testing

The last section of the develop plan called for the design of the commercial system and pilot testing. First, different subsections of the system were modeled to determine the proper designs. These included thermal gradient modeling of the filter mounted assemblies, gas flow modeling of the backpulse system, and gas flow modeling of the dirty side of the filtration system (reentrainment). Then, the entire commercial system design was put together and analyzed from the perspective of manufacturing, transportation, operation, reliability, maintenance, and capital and operating cost. The results of the Product Development Program to date indicate that the technology is now ready to be pilot tested using production scale filters in a system that represents the commercial system design.

Results

CRADA Objectives

As an example of the type of work that was performed in the Product Development Plan, the 300 hour filtration evaluation will be described. This test was sponsored, in part, by a CRADA between CeraMem and DOE METC, No. 93-012. Aspects of the project were subcontracted to the University of North Dakota’s Energy and Environmental Research Center, Foster Wheeler (formerly Ahlstrom Pyropower), and The Pennsylvania State University.

The purpose of this test was to evaluate the filtration performance of CeraMem filters on “live” flue gas at temperatures similar to those in PFBC’S. The objectives of the test to be discussed in this paper areas follows:

- To demonstrate that the CeraMem filters could be cleaned effectively for 300 hours while operating on “live” particulate-laden flue gas.
- To evaluate filter operating parameters such as filter cleaning frequency.
- To investigate ash-to-filter chemical interactions.

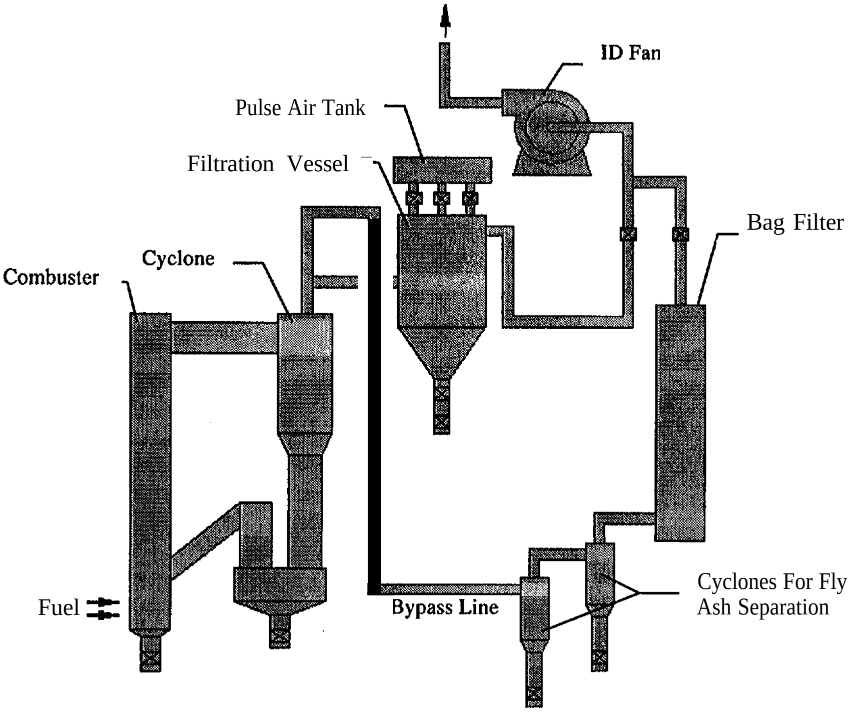

Figure 3. CRADA Filtration Vessel

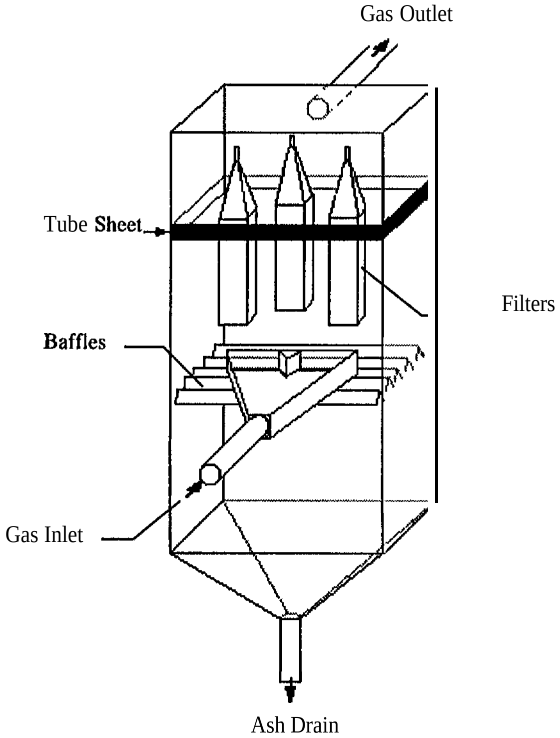

Figure 4. Circulating Fluidized Bed Combustor System Used in CRADA Testing

CRADA Materials and Equipment

The filters in this test were identical to commercial filters except that the cross section was reduced to fit the quantity of gas flow through the test facility. The filters had an 84 mm square cross section and 381 mm length with a filtration surface area of 0.62 m^2. The filters were sealed into housings using a similar approach to the design used in the commercial housing design. One end of the housing was flanged in order to attach the housed filter to the filtration system tubesheet.

Filters were tested three at a time in a single filter vessel (Figure 3) which was attached to a bench scale atmospheric circulating fluid bed combustor (CFBC; Figure 4). The test vessel was comprised of three sections. The upper section above the tube sheet contained the venturis, pulse equipment, and clean gas outlet. The middle section held the tubesheet to which the filters were attached. The lower section located below the filter inlets contained the flue gas inlet, baffles, and ash hopper.

The filtration vessel design and resulting flue gas flow was the result of modeling conducted at DOE METC. The approach to the design was to ensure uniform gas flow to the filters while minimizing any chance of ash reentrainment back into the gas flow entering the filters. The flue gas flow entered the side of the vessel from the CFBC. The gas was allowed to expand to reduce its velocity and promote drop out of large particles from the flue gas stream. After entering the vessel, the flue gas turned ninety degrees upward to enter into the bottom of each of the three filters, The flue gas was filtered as it proceeded up through the filters and then exited the vessel by flowing through the venturis and then out the clean gas outlet. During backpulsing, jets of gas and particles were sent through the arrow baffles located just below the flue gas inlet. The intent of the baffles was to minimize ash reentrainment into the inlet flue gas flow. Once below the baffles, the backpulse gas velocity was reduced due to the volume of the vessel and the particles were allowed the settle out of the gas stream. Ash was removed from the hopper via a slide gate valve.

The backpulse system was fabricated to allow individual pulsing of each filter. Compressed air at 6.5 bar was pulsed by fast acting valves through dedicated nozzles into venturis located over each filter. The combination of the gas pulsed into the venturi and gas flow induced by the venturi design resulted in a pressure differential across the filter large enough the dislodge the ash from the filter.

The entire vessel was instrumented with fast response pressure transducers and thermocouples to allow for evaluation of pressures and temperatures during the testing, All data was acquired through two independent data acquisition systems. One system was used to record the combustion and emission data at slow rates. The second system acquired filter vessel data (temperature or pressure readings) at either slow rates during normal operation or at high rates during backpulsing. This second system also triggered the backpulsing sequence.

The test facility fired a mixture of high sulfur bituminous coal (Illinois No. 6; 9.7% ash) and industrial grade limestone (Iowa Industrial Lime No. 1) that had been premixed prior to being introduced into the combustor. The resultant fuel and limestone mixture gave a Ca/S molar ratio of 2.

CRADA Operating and Test Procedures

The filter testing was conducted in two 150 hour tests. During the first test, three filters were tested. After completing the first test, one filter was removed and a fourth new filter was installed, After completing the change out, a second 150 hour test was conducted. At the end of the 300 hour test period, two filters had been exposed to 150 hours of operation while two others had been exposed to 300 hours of operation.

The test facility operated very well during the two 150 hour tests. The fuel feed rate was approximately 5,5 kg/hr resulting in an average bed temperature of 870°C. The combustion efficiency was about 99,8% for each run. The resulting flue gas (dry basis) had about 17% CO_2 and 4% O_2.

The filter operating conditions for the two 150 hour tests were very similar. The filters were operated at a temperature of approximately 863°C and a total gas flow of about 39.7 Nm^3/hr which resulted in a face velocity of about 2.24 cm/s. The baseline pressure drop across the filters at the start of the tests was about 25 mbar. The filters were automatically regenerated at 40 mbar differential pressure in each test. The number of pulses per cleaning cycle was one pulse per filter per cleaning cycle for the first test and two pulses per filter per cleaning cycle for the second test.

During each 150 hour test, ash samples were taken at the filter vessel inlet and outlet by personnel from the University of North Dakota's Energy and Environmental Research Center, Samples were obtained using EPA Method 5 taking approximately 1% of the total flow.

In order to measure the thermal shock that the clean side of the filters experienced during the very rapid backpulse process, optical pyrometers were used. Three high-speed, two-color, fiber optic pyrometer systems were used to measure the filter surface temperature at the center, edge, and corner of one filter during backpulsing. These tests were performed at approximately 600°C due to system limitations prior to the systems being completely debugged, However, these measurements gave an indication of the type of thermal shocks that the filter materials were exposed to without the effects of instrument lag experienced by other thermal sensors.

After the tests were completed, the midsections of the two filters that were exposed to 300 hours of operation were sent to The Pennsylvania State University for mechanical property characterization, Flexural strength specimens about 5 cm in length were machined from the filter samples, Two different types of specimens were prepared for mechanical testing. A "clean-side" specimen was prepared where the maximum tensile stress during the flexural strength test was generated along a surface which was not exposed to particulate. A "dirty-side" specimen was prepared where the maximum tensile stress during the flexural strength test was generated along a surface which was exposed to particulate. Four point bend tests were performed on approximately 10 specimens at each of three different temperatures.

CRADA Results

Figure 5 shows a plot of filter pressure drop versus time for a selected 50 hour section of the first 150 hour test. As indicated above, the backpulse was triggered at 40 mbar after an increase in filter pressure drop from the initial 25 mbar. Overall, the backpulse cleaning resulted in cleaned filter pressure drops of about 28.6 mbar and 29.9 mbar for the two tests respectively after 150 hours of operation, In both cases, the initial duration between pulse cleanings was about 45-50 minutes at the start of the test and decreased to 30 minutes at the end of the test, The slight increase in filter pressure drop and decrease in filtration cycle time are due to filter conditioning under the test conditions. In this test, the use of double pulses was not more effective than a single pulse in maintaining a lower clean out filter pressure drop.

Figures 6 and 7 show plots of filter permeance versus time for both 150 hour tests, Permeance is defined as follows:

$$k_{20"C} = \frac{v \times \mu}{dP \times \mu_{20^\circ C}}$$

where

$k_{20^\circ C}$ = permeance, cm/s-mbar at 20”C
v = filtration face velocity, cm/s
μ = dynamic viscosity at test conditions
dP = pressure drop, mbar
$\mu_{20^\circ C}$ = dynamic viscosity at 20”C

The use of permeance normalizes the gas filter pressure drop to face velocity and temperature which may vary during an actual test.

In both Figures 6 and 7, the permeance is approaching a constant value. This indicates that the filters are going through an initial conditioning which may be the result of various ash-to-filter interactions. Over a long filtration run, the backpulse cleaning mechanism would continue to clean the filters and maintain the permeance at a value slightly less than that indicated after each 150 hour run,

Table 1 shows the particulate sampling information obtained by the University of North Dakota. For both tests, the dust loading entering the filter vessel was between 1.8 and 1.9 g/Nm^3. The dust loading exiting the filter vessel was much smaller at 0.0025 g/Nm^3 and 0,0011 g/Nm^3 for the two tests respectively. While the outlet dust loading is well below the target maximum concentration for gas turbine operation, the effective particulate capture efficiencies do not appear to be overly high. Analysis of the outlet ash collected during the tests indicated that a significant percentage of the outlet materials was from system contamination. As a result, the actual filtration efficiencies of the filters should be much higher than those reported.

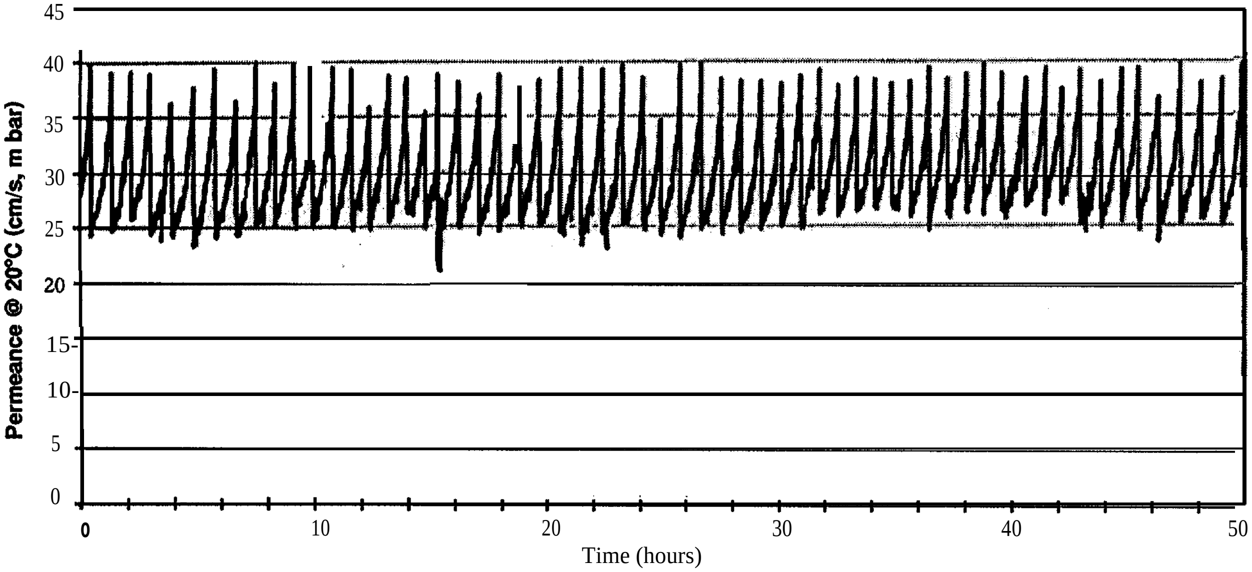

Figure 5. Selected 50 Hours Interval of Filter Pressure Drop Versus Time from First Test

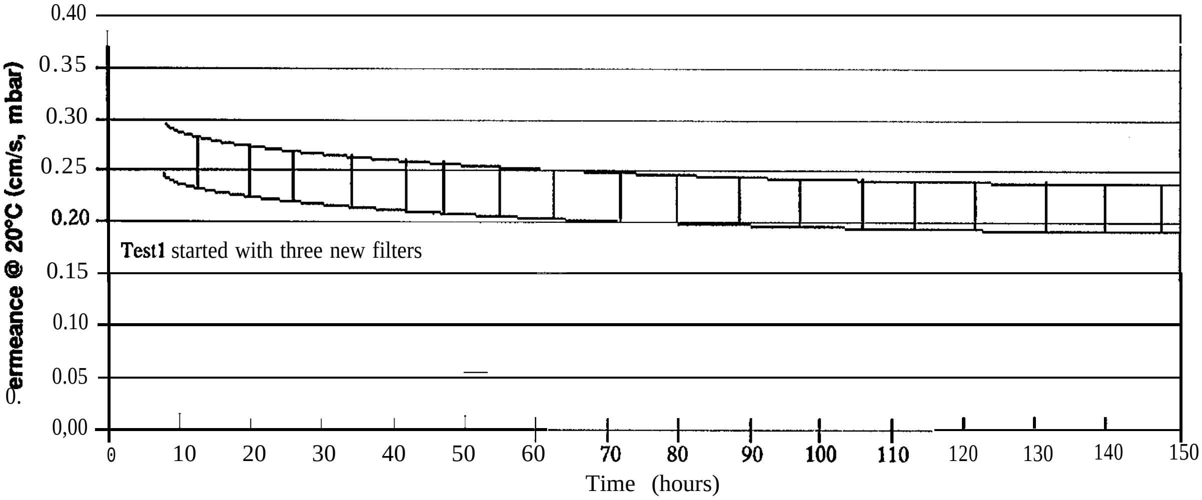

Figure 6. Permeance Versus Time for First 150 Hour Test

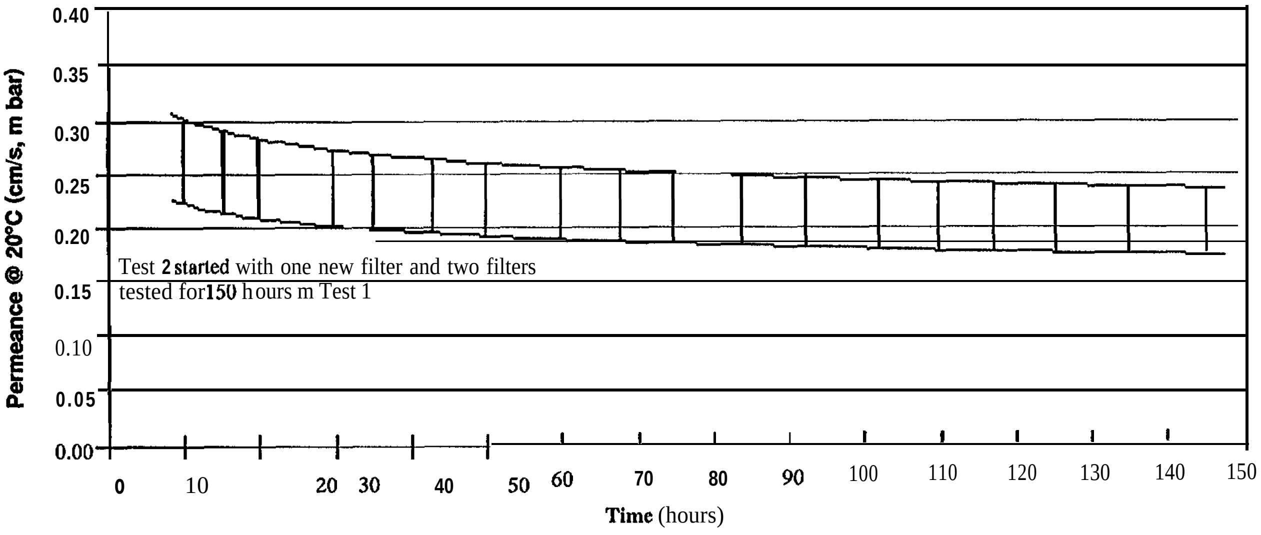

Figure 7. Permeance Versus Time for Second 150 Hour Test

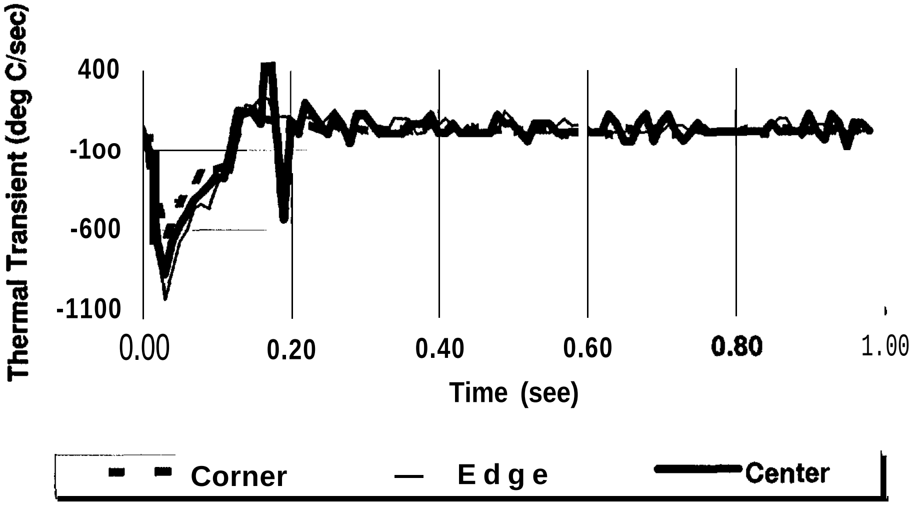

Figure 8. Gas Filter Termal Shock During Backpulse at 600°C

Table 1. Particulate Removal Efficiency as Measured in Tests 1 and 2

Parameter	Test Number 1	Test Number 2
Inlet Loading	1.88 g/Nm^3	1.83 g/Nm^3
Outlet Loading	0.0025 g/Nm^3	0,0011 g/Nm^3
Efficiency	99.87%	99.94%

Figure 8 shows the type of thermal shock the clean face of the filter is exposed to during operation, Thermal shocks approaching 1000”C per second were measured using the optical pyrometry equipment. It should be noted that these thermal shocks had no effect on the cordierite monolith support material due to its very low thermal expansion coefficient,

Table 2 shows the results of the flexural strength tests as a function of temperature. It is clear that the filter materials were as strong after 300 hours as they were at the start of the test, In addition, it makes no difference whether the surface of maximum tensile stress during the flexural strength test was exposed directly to the particulate-laden flue gas or not. As a result, it can be concluded that ash and alkali contaminants which impinged on the filter surfaces had not interacted with the filter materials so as to change their inherent strength. In addition, the thermal shocks that were experienced by the filter during each backpulse sequence had no effect on filter strength.

Table 2. Filter Flexural Strength as a Function of Temperature After Testing for 300 Hours

Temperature	Average Flexural Strength (MPa)		
	Unexposed	Clean Side	Dirty Side
20°c	10,1 +/- 0.4	10.9 +/- 0.7	10.4 +/- 0.7
800°C	10,2 +/- 1.3	11.3 +/- 1.8	12.3 +/- 2.2
870°C	No Data	10.7 +/- 1.5	12,5 +/- 1.0

CRADA Conclusions

Four filters were successfully tested over the course of a 300 hour test using “live” flue gas at 870°C that was produced by an atmospheric circulating fluidized bed combustor. Specifically, the filters were able to be cleaned over the time period with a small increase in filter resistance attributed to filter conditioning. Particulate removal was considered to be high (> 99.94%) since a significant amount of material seen downstream was concluded to have been contamination. Thermal shock evaluations indicated that the filters were exposed to gradients approaching 1000°C/s based on optical measurements, However, even with exposure to this thermal shock and ash and alkali at high temperature, there was no indication that the filter material degraded. Furthermore, it was concluded that long term testing under process conditions of commercial interest was a necessary next step in the development of the technology.

Benefits

Based on the Product Development Program tasks completed to date, several benefits can be seen in this technology as compared to candle filter technology. These are described as follows.

1, The honeycomb monolith structure is less prone to catastrophic failure, The combination of advanced monolith support material microstructure, relatively equiaxed honeycomb structure, and rugged mounting design make it very unlikely that the filters will catastrophically fail and leave a gapping hole through which particulate-laden flue gas can pass.
2. Compared to some candle filter materials, the strength of the CeraMem filter does not change as a function of temperature. Also, due to the high temperature corrosion resistance of the materials, the filters can withstand high temperature oxidizing conditions better than some candle filter materials.
3. The thermal shock resistance of the cordierite monolith is excellent due to its phase composition and advanced microstructure.
4. The present CeraFilter commercial filtration system design has a higher packing density (surface area per unit volume of vessel and surface area per vessel) than candle filter systems by a minimum of 25%. As a result, not only are there fewer vessels required to process a particular gas stream, but the quantity of auxiliary equipment (e.g., compressors), structural steel, and plan area are significantly reduced.
5. Once demonstrated, the present CeraFilter commercial filtration system design will be a minimum of 25% less costly over a 20 year life cycle as compared to candle filter systems.

Future Activities

The status of the overall development program is that the technology is ready for pilot testing. CeraFilter is presently pursuing opportunities to conduct pilot testing over long periods of time (2,000 to 10,000 hours) on advanced combustion power systems of reasonable size so as to make them representative of the full size commercial system.

Acknowledgements

The authors would like to acknowledge the support of DOE METC under SBIR Grant Number DE-FG02-90ER80896 and Mr. Norman Holcombe who was the Contracting Officer's Representative, The period of performance for this grant was May 1991 to March 1996. The authors would also like to thank DOE METC for the CRADA support (Number 93-012) and DOE's project manager, Mr. Douglas Straub. In addition, the authors would like to acknowledge the subcontractors who worked on the CRADA project: Dr. K. Katrinak and Ms. T. Watne of the University of North Dakota's Energy and Environmental Research Center, Mr. J. Tuncay of the Foster Wheeler Development Corporation, and Prof. Richard Tressler at The Pennsylvania State University.