

# Iron Aluminide Hot Gas Filter Development

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## 1.0 Introduction

The development of advanced, coal fired, power generation systems such as pressurized fluid-bed combustion (PFBC) and integrated gasification combined cycles (IGCC) is an important part of the future energy picture for the United States and the world. These technologies can provide economical power generation with minimal environmental emissions and high efficiency. These advanced power generation projects are, however, dependent on the development of durable economical high temperature filter systems.

Currently high temperature filter systems are in the demonstration phase with the first commercial scale hot filter systems installed on IGCC units and demonstration units of PBFC systems. These filters are mostly ceramic tubes or candles. Ceramic filter durability has not been high. Failure is usually attributed to mechanical or thermal shock.

For IGCC the major problem associated with the use of ceramic filters is their lack of resistance to cracking due to mechanical loads. One possible solution to this problem is the development of sintered metal filters (which are more resistant to cracking than ceramic filters) which can withstand the hydrogen sulfide laden, high temperature gases of these systems. The purpose of this project is to develop crack resistant, corrosion resistant sintered metal filters of iron aluminide suitable for application in advanced power processes. The goal is to develop filters which will provide at least several years service in advanced power gasification applications without a substantial temperature penalty.

## 2.0 Objectives

The overall objective of this project is to commercialize weldable, crack resistant metal filters which will provide several years service in advanced power generation processes. These filters will be used to remove particulates from the gas stream prior to entering a turbine.

Three objectives of the previous portion of the project were to (1) develop filter media from corrosion resistant iron aluminide alloys, (2) develop manufacturing processes to make iron aluminide filters and (3) use a "short term" exposure apparatus, supported by other tests, to identify the most promising candidate (alloy plus sintering cycle).

The current phase objectives are to demonstrate longer-term corrosion stability for the best candidates followed by the production of fifty filters.

### **3.0 Approach**

Pall Corporation has taken the approach of developing sintered metal filter materials suitable for use in advanced power production applications. The focus of this project is to develop Iron aluminide for use in gasification environments. Specifically, three alloy compositions were chosen and modified after reviewing the relevant literature (1-7). The primary considerations for the alloys were resistance to spalling and corrosive attack in a reducing environment containing sulfur and chlorine and acceptable mechanical properties including ductility and tensile strength.

Two compositions were selected for corrosion testing in a simulated IGCC atmosphere. A Fe<sub>3</sub>Al with 2% chromium (FAS) and another with 5% chromium (FAL) were chosen based on their performance in previous short term corrosion testing. The test samples were prepared using standard techniques for manufacturing seamless porous media by the Pall Corporation.

### **4.0 Project Description**

This project is divided into three major tasks: The first task was to develop manufacturing methods for Iron Aluminide. This task was largely completed and reported on in our previous paper at the previous DOE conference. The second task was to assess the corrosion resistance of the iron aluminide in simulated gasification atmospheres with both short and long term exposure tests. The third task (optional) is to produce 50 commercial scale filters.

This paper will focus on the long-term exposure of the second task.

#### **Long Term Corrosion Testing**

Long-term corrosion testing of the two heat-treated iron aluminide compositions was performed in a simulated IGCC atmosphere containing 78,300 ppm H<sub>2</sub>S at 925 °F and one atmosphere. This testing was used to identify trends in the corrosion of the iron aluminide media. Previous short term testing provided excellent results however, trends were not evident.

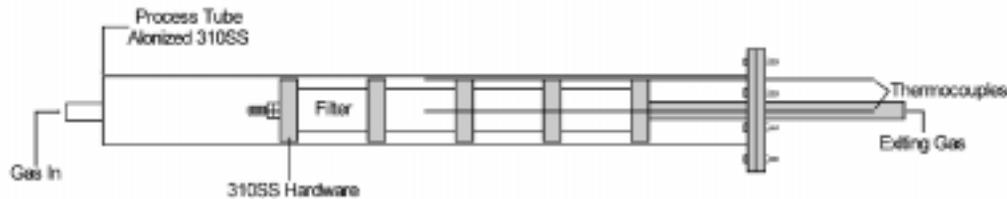
##### **4.1.1 Heat Treatment of Elements**

The filter samples were heat treated prior to exposure to the simulated IGCC gas stream. Heat treatment allows the formation of an alumina layer. The alumina layer formation on a porous body would take too long in reducing conditions. Each area of the filter would be "competing" for oxygen in the reducing conditions.

### 4.1.2 Corrosion Test Apparatus

The same test stand was used that was discussed in previous paper. The stand allows the samples to be exposed to flow through conditions simulating actual service.

The filter samples were held in place between 310 stainless steel hardware with a tie rod to make a “flow through” assembly (see Figure 1). The media was not welded to the hardware. The filter string was disassembled at regular intervals for non-destructive testing. The filters were rotated in the filter string as is common practice in corrosion testing.



**Figure 1. Schematic of Process Tube Furnace for Long Term Exposure Testing**

### 4.1.3 Test Atmosphere Components and Experimental Approach

The atmosphere consisted of a mixture of hydrogen, carbon monoxide, methane, hydrogen sulfide and steam with sodium chloride, potassium chloride and hydrochloric acid. Table I lists the operating conditions for representative oxygen blown (Tampa Electric) and air blown (Sierra Pacific) IGCC atmospheres at system pressure. This table also lists the test atmosphere (without nitrogen) that was used, at approximately one atmosphere, to simulate both the oxygen blown and the air blown installations. The composition for this simulation atmosphere was determined by Oak Ridge National Laboratory (ORNL). The face velocity chosen was 0.5 feet per minute in forward flow. During the thermal pulsing the velocity was 18 feet per minute.

Hydrogen, Carbon Monoxide, Carbon Dioxide and Methane were dispensed from individual pressurized cylinders. Hydrogen sulfide was dispensed from a tank (liquid phase). Provisions were made to measure the hydrogen sulfide levels before and after the gas passed through the filter string. Each day the H<sub>2</sub>S level was monitored at the inlet and at the outlet of the furnace tube. To measure the H<sub>2</sub>S levels a Toxic Gas Detector Model 8014KA (Matheson-Kitagawa) was used. The H<sub>2</sub>S inlet and outlet ports were hooked up in a tee, this allowed the gas to be flowing while the H<sub>2</sub>S was being measured. The hydrogen sulfide level outlet was kept within 15% of the target level. A reservoir filled with DI water plus NaCl, KCl, and HCl supplied the water and chlorides to the test stand.

The furnace atmosphere flowed from the outside to the inside of the test filters which simulated use. The simulation gas was mixed in the process tube, flowed through the filters and then exited the furnace. Each run exposed four samples at one time.

The simulated exposure conditions of 78,300 ppm H<sub>2</sub>S at 925 °F and one atmosphere have been calculated (by Peter Tortorelli, ORNL) to be equivalent to 0.72 mol % H<sub>2</sub>S at 26.1 atmospheres in an oxygen blown gasifier.

#### **4.2 Material and Property Analysis**

Each of the exposed samples were analyzed and compared to the results of control materials. The analyses completed on each sample are shown below:

##### **Material Analyses**

- Carbon/Sulfur
- Strength
- Ductility
- Porosimetry
- SEM/EDS

##### **Property analyses**

- Mass
- Air DeltaP
- Bubble Points

The air delta P was measured in inches of water across the filter media at a flow of 28 acfm/ft<sup>2</sup>. For bubble point tests The samples were wet in and submerged approximately half inch below the surface of Filmex-B (denatured ethyl alcohol) prior to testing. Stoppers were placed in the open ends of the samples. Air pressure inside the element was gradually increased. The pressures at which the 1st and 10th bubbles occurred were recorded. The first bubble point is the pressure at which a bubble of air escapes from the largest pore in the sample: the first bubble point can be correlated to the absolute filter efficiency. The 10th bubble point was compared against the 1st bubble point to judge the uniformity of the pore size.

The open bubble point was also recorded. The open bubble point is an indication of the pressure required to pass a specified quantity of air (1 scfm/ft<sup>2</sup>) with the element wet in Filmex and relates by experience to the average pore size.

Table I

Representative IGCC Atmospheres and a Simulated Atmosphere for Exposure Testing

Types of Atmosphere	Oxygen Blown Tampa Electric	Air Blown Sierra Pacific	Simulated* Atmosphere (w/o Nitrogen) w/ chlorides
Temperature °F	900 - 925	1000 - 1050	Tampa - 925 Sierra - 1050 See Table II
Pressure	400 psia 26.1 atmosphere	272 - 275 psia	~ 1 atmosphere
<u>Component</u>	<u>Value - Mole %</u>	<u>Value - Mole %</u>	<u>Value - Mole %</u>
CO	40.36	28.89	34.1
H <sub>2</sub>	28.20	14.57	31.4
CO <sub>2</sub>	10.34	5.44	15.7
H <sub>2</sub> O	14.16	5.50	9.2
CH <sub>4</sub>	0.15	--	0.92
Ar	0.94	0.60	--
N <sub>2</sub>	5.13	48.65	--
COS	0.02	--	--
O <sub>2</sub>	0.00	0.00	0.00
H <sub>2</sub> S	0.63**	0.03**	7.83
HCl	NA	NA	80 ppm
NaCl	NA	NA	2x*** 2 ppm
KCl	NA	NA	2x*** 5.5 ppm

\* Corresponds with oxygen blown Tampa Electric, Equilibrated at 1300°F, at 1 bar with no nitrogen.

\*\* Upstream of final desulfurization which is expected to lower H<sub>2</sub>S to 0.003% (30 ppm)

\*\*\* Amount added

**Note:** Temperatures and pressures supplied by METC.

## 5.0 Results

As can be seen in figure 2, the change in mass of Iron aluminide over time of exposure remained very low over the 1500 hours of testing with less than 0.45% change for most of the samples. A majority of the mass gain occurred within the first 500 hours of exposure. After approximately 500 hours of exposure the rate of mass gain slowed considerably.

The change in pressure drop of iron aluminide porous media appears to change linearly with exposure (Figure 3). This is not uncommon for porous material exposed in harsh environments. The change in pressure drop may appear to be substantial, but in an operating system the pressure drop across the media is very small, with most of the pressure drop occurring in the dust cake. Therefore, large changes in pressure drop through the media can be seen with very slight changes in the overall system pressure drop. In addition, we have seen pressure drops very close to “as new” condition from physical cleaning the elements (i.e. removal of the elements from the system and backflushing with an appropriate chemical)

SEM evaluations of the media exposed for 336 hours showed no serious sulfidation. SEM cross sections (FAS alloy only) shows that the increase in mass is from iron sulfide forming on the upstream surface. The iron sulfide formation is not from the base metal and is believed to be from residual iron left on the exterior of the alumina scale during heat treatment. A comparison of the sample exposed for 500 hours (Figure 5) to the 1000 hour (figure 6) and 1500 hour samples (Figure 7) reveals that the amount of iron sulfides forming on the iron aluminide media is not increasing after 500 hours of exposure. This agrees with the mass gain curves.

In an attempt to project the mass gain of the filter element over its desired lifetime, we extrapolated (using two different extrapolation techniques) the mass gain using data from the exposed elements. Both extrapolations assume that there is no marked change in corrosion mechanisms of the material over the life of the filter. A linear extrapolation was done using a best fit of the points forming the plateau (500 – 1500 hours) after the initial mass gain. This extrapolation technique is a crude method to use for a solid material, however, for a porous material that has demonstrated a marked increase in mass from the first portion of the exposure, this type of extrapolation may be useful. The linear curvefit projected a mass gain of 2.4% after 24,000 hours of service.

A parabolic fit was done on the entire curve that allows a more traditional estimation of the mass gain of the porous alloy. This fit agrees with the final three points of the 1500 hour exposed samples (FAS sample #2). This curvefit predicted a mass gain of 1.8 % over 24,000 hours of exposure.

These predicted mass gains are miniscule and would be undetectable on a filter in actual service due to the weight of the dust cake.

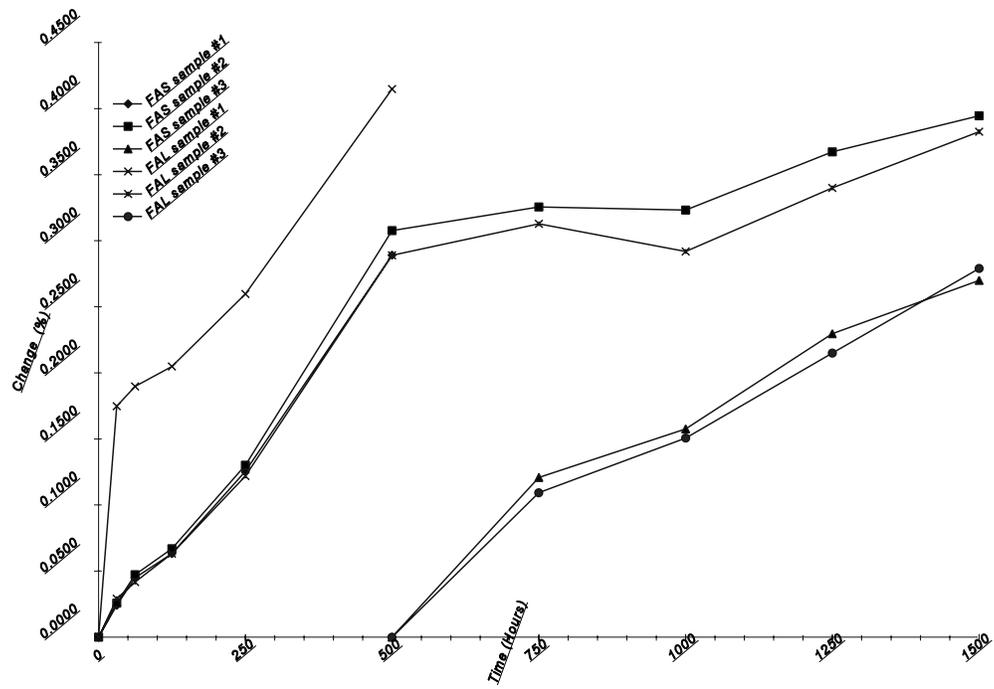


Figure 2: Change in mass of iron aluminide porous media exposed to 78,300 ppm H<sub>2</sub>S at 925° F and one atmosphere.

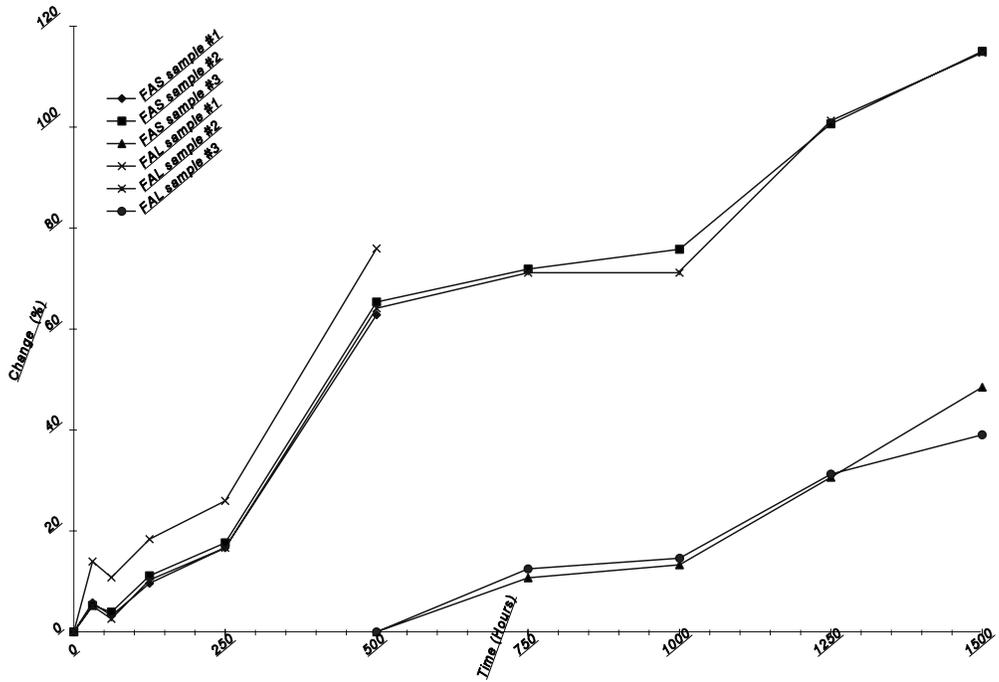
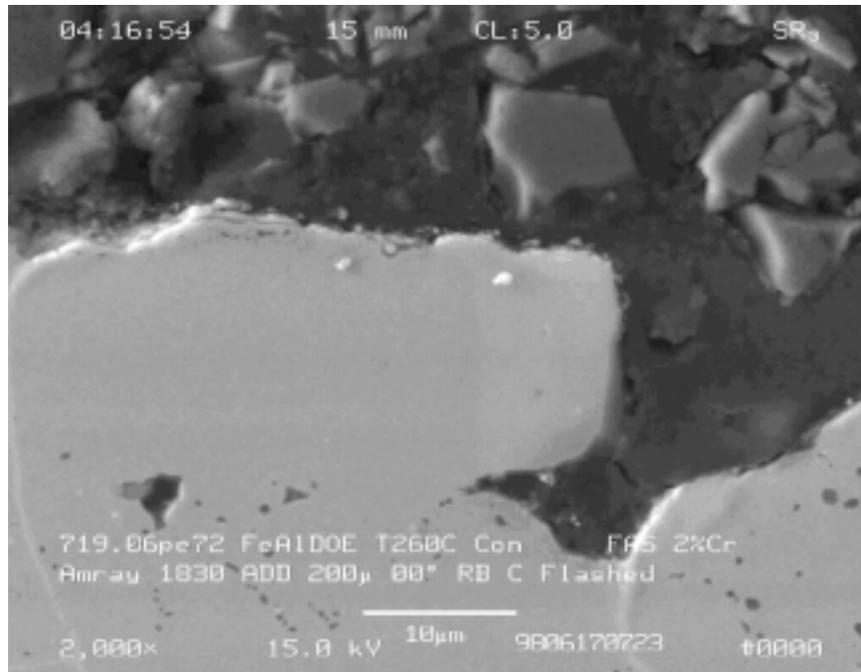
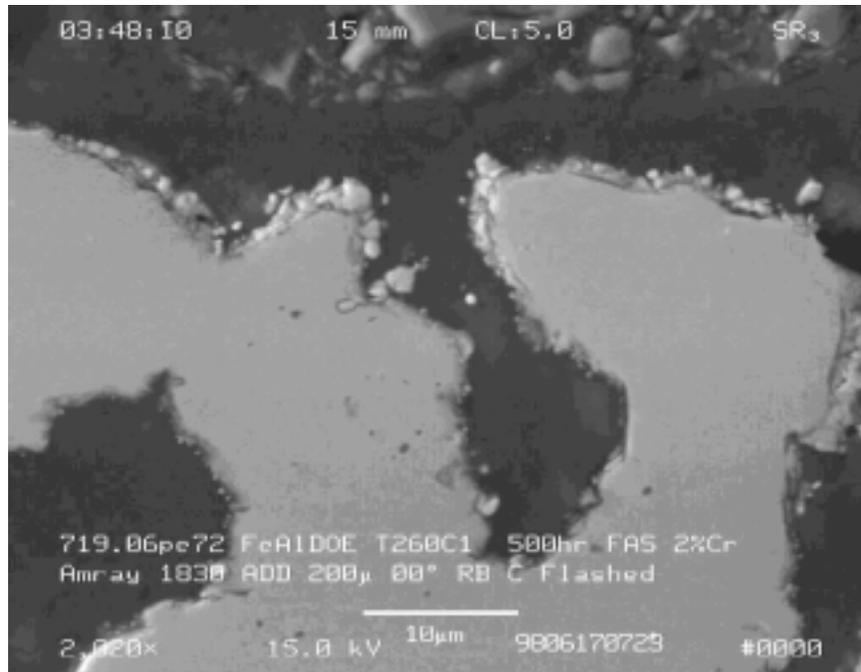


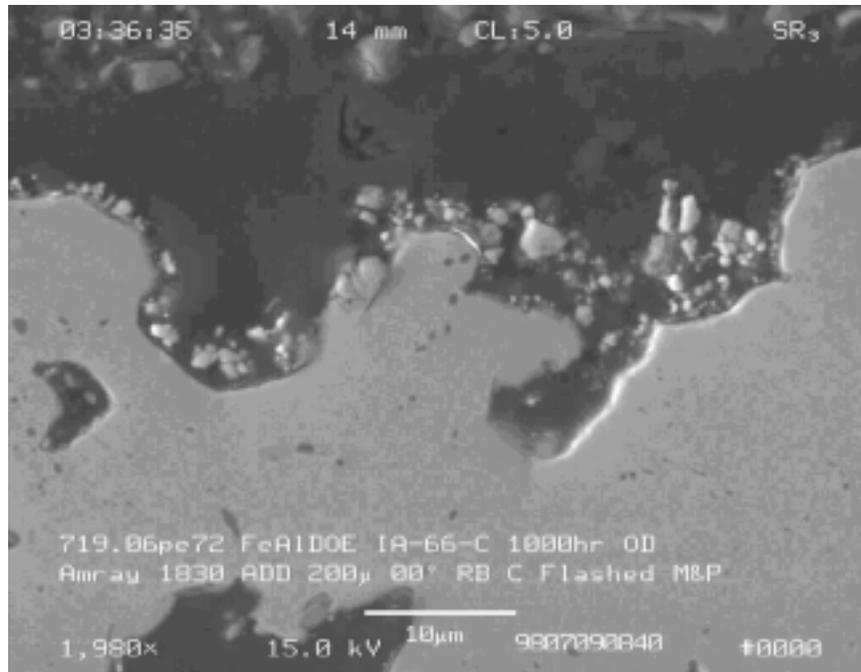
Figure 3: Change in pressure drop of iron aluminide porous media exposed to 78,300 ppm H<sub>2</sub>S at 925° F and one atmosphere.



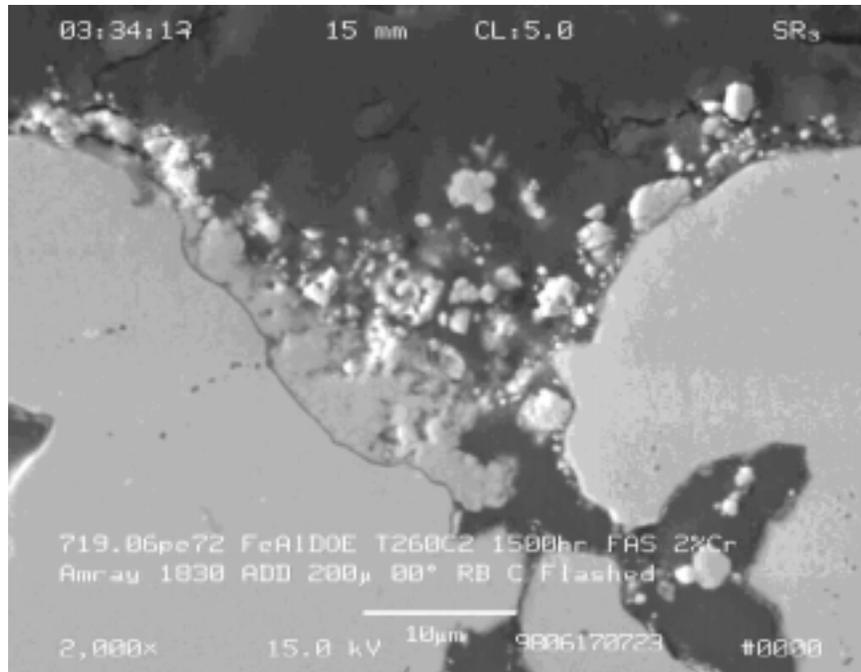
**Figure 4: Upstream edge of FAS control sample. Robinson backscatter. Typical iron aluminide. Dark inclusion are alumina from the water atomization. (2000X)**



**Figure 5: Upstream edge of cross-sectioned FAS sample exposed for 500 hours. Robinson backscatter. Iron sulfides on the surface of the particles. Base metal is not attacked. (2000X)**



**Figure 6: Upstream edge of cross-sectioned FAS sample exposed for 1,000 hours. Iron sulfides on the surface of the particles. Approximately the same amount of sulfides as the sample exposed for 500 hours (Figure 5). Base metal is not attacked. Robinson backscatter. (2000X)**



**Figure 7: Upstream edge of cross-sectioned FAS sample exposed for 1,500 hours. Iron sulfides on the surface of the particles. Approximately the same amount of sulfides as the sample exposed for 500 hours (Figure 5). Base metal is not attacked. Robinson backscatter. (2000X)**

The mechanical properties of the FAS media were unchanged within the accuracy of the test methods. The samples tested for 1500 hours lost 7.6% strength and 9.8% ductility. Experience has shown that iron aluminide media usually has a loss of strength and ductility after short exposures in simulated environments and actual service. The loss in strength and ductility occurs early in service and may not continue to degrade the material properties.

## **Conclusions**

Pall has developed a crack resistant metal filter element that is suitable for IGCC application. The filter material developed has surpassed all the performance targets established at the outset of this PRDA.

This filter will initially be available in the 2%Cr alloy. The 5% Cr alloy may become commercially available, however additional testing is required to determine its advantage and disadvantages when compared to the 2%Cr alloy.

Production of the 2% Cr alloy is currently being moved to manufacturing from the development laboratories. This represents one of the last steps in the commercialization process. We expect that the filter material will be commercially available within 6 months.

The life expectancy is estimated to be greater than 24,000 hours at typical IGCC conditions. This is based on a corrosive failure. Exposure of iron aluminide in actual service conditions will clarify what factors will contribute to its failure. Once the factors are known a better estimation of filter life can be made.

## **Application**

The iron aluminide media developed under this program has been specifically targeted for application in reducing environments, which contain hydrogen sulfide and chlorides, i.e., coal gasification. The targets originally set were application at 1250°F at H<sub>2</sub>S levels up to 3000 ppm<sub>v</sub>.

From the results of the tests to date, it appears that the material will provide long term operation at H<sub>2</sub>S levels substantially greater than those originally targeted.

This media was developed primarily for one reason. Our preliminary costing indicate that a full scale filter system utilizing iron aluminide media can have a capital cost 30% less than one designed exclusively for ceramic filter elements. This is a significant economic driver to pursue commercial application of iron aluminide filter elements in gasification systems.

In addition, with the sintering and manufacturing techniques developed in this program, we believe that it is possible to develop sintered metal filter elements which may be suitable for PFBC application. Initial discussions with ORNL indicate that 5% Cr iron aluminide may be suitable for application in PFBC. There are other alloys that are also promising, which may be sinterable using the techniques developed.

## **8.0 Future Activities**

Pall has recently been given a contract extension to complete long term exposure testing of the candidate Iron aluminide media. This testing will take place this fall under simulated gasification conditions.

In addition, DOE has the option of directing Pall to manufacture 50 commercial scale iron aluminide filters for field testing in actual gasification operation.

## 9.0 Acknowledgments

The authors acknowledge the advice and encouragement of our METC Project Manager Ted McMahon. Special thanks are due to several individuals at Pall including Steve Geibel (PED) for guidance, to Joe Puzo (MMD) for help in the spinning and vacuum sintering of seamless cylinders and to Keith Rekczi (PED), a team member, for running experiments and for help in upgrading the exposure apparatus. Special thanks are due as well to several staff members at Oak Ridge National Laboratory; these include Peter Tortorelli and Jack de Van (Ret.) for calculating equilibrium atmospheres.

## 10.0 Contract Information

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