Moving Granular Bed Filter Development Program

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Introduction

For coal-fired power plants utilizing a gas turbine, the removal of ash particles is necessary to protect the turbine and to meet emission standards. Advantages are also evident for a filter system that can remove other coal-derived contaminants such as alkali, halogens, and ammonia. With most particulates and other contaminants removed, erosion and corrosion of turbine materials, as well as deposition of particles within the turbine, are reduced to acceptable levels. The granular bed filter is suitable for this task in a pressurized gasification or combustion environment.

Objectives

The objective of the base contract was to develop conceptual designs of moving granular bed filter (GBF) and ceramic candle filter technologies for control of particles from integrated gasification combined cycle (IGCC), pressurized fluidized-bed combustion (PFBC), and direct coal-fueled turbine (DCFT) systems. The results of this study showed that the GBF design compared favorably with the candle filter.

Three program options followed the base contract. The objective of *Option I, Component Testing,* was to identify and resolve technical issues regarding GBF development for IGCC and PFBC environments. This program was recently completed. The objective of *Option II, Filter Proof Tests*, is to test and evaluate the moving GBF system at a government-furnished hot-gas cleanup test facility. This facility is located at Southern Company Services (SCS), Inc., Wilsonville, Alabama. The objective of *Option III, Multicontaminant Control Using a GBF*, is to develop a chemically reactive filter material that will remove particulates plus one or more of the following coal-derived contaminants: alkali, halogens, and ammonia.

Approach

The GBF was developed through low-pressure, high-temperature (1,600 °F) testing in the late 1970s and early 1980s. Collection efficiencies over 99 percent were obtained. In 1988, high-temperature, high-pressure (HTHP) testing was completed at New York University (NYU), Westbury, New York, using a coal-fired pressurized fluidized-bed combustor. High particulate removal efficiencies were confirmed by the fact that both New Source Performance Standards (NSPS) and turbine tolerance limits could be met (Wilson, 1989).

The early scale-up work of the GBF indicated potential limitations due to size, cost, and mechanical complexity. These limitations were addressed in the current program by using the information gained from the filter development up through the NYU test program to reassess the commercial approach (Wilson et al., 1992). Figure 1 shows a GBF designed for a 100-MWe Kellogg-Rust-Westinghouse (KRW) air-blown fluidized-bed gasifier. The filter has an inside diameter of 14 ft with a nominal bed depth of 5 ft.

Dust-laden gas enters the filter through an inlet duct, which brings the gas and dust to the gas filter medium interface. The gas flows downward initially, turns and flows across the filter medium, and then turns upward to flow counter-current to the downward moving filter medium. As the gas and dust pass through the filter medium, dust is removed. Dust and filter media are removed at the bottom cone of the filter. A pressurized pneumatic lift pipe moves the filter medium for return to the filter. After separation of the filter medium, the transport gas and dust are cooled in a recuperative heat exchanger and pass through a pressurized baghouse, which removes the collected dust. The cleaned transport gas pressure is boosted 15 psi through a blower, reheated in the recuperative heat exchanger, and returned to the bottom of the lift pipe. The cleaned filter medium from the disengagement vessel at the top of the lift pipe flows by gravity back into the top of the filter.
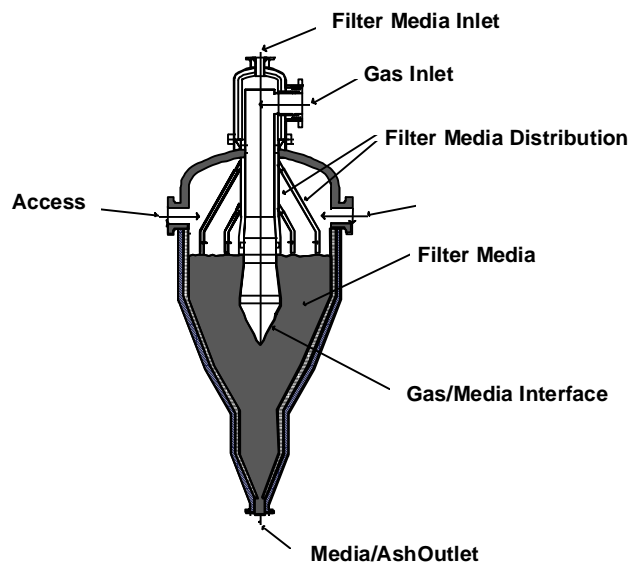

Figure 1. Granular Bed Filter for 100-MWe KRW (Air) Gasifier

Project Description

The status of three GBF projects is reported in this paper: (1) the recently completed GBF component development program (Option I), (2) the development of multicontaminant control filter material (Option III), and (3) impending filter proof tests (Option II).

GBF Component Development Program

Technical issues that are associated with the development of a GBF for the control of particulate in IGCC and PFBC environments were addressed. Some of the technical issues are due to problems encountered during testing of a GBF at NYU. Other issues are a result of the redesign of the GBF to make it more commercially competitive. The issues are prioritized based on the seriousness of the issues and the difficulty in resolving them. Some issues are amenable to being settled in a component test facility, while others will have to be dealt with in the U.S. Department of Energy's (DOE's) Power Systems Development Facility (PSDF) located at an SCS facility in Wilsonville, Alabama.

A test plan defined a component test program to address the following issues:

- Effect of filter cone angle and sidewall materials on medium flow and ash segregation
- Maximum gas filtration rate
- GBF media issues
- Filter pressure drop.

A 3.5-ft-diameter split filter was used with a transparent cross section that allowed visual examination of filter media characteristics. Two different-sized filter arrangements were used for the evaluation, with different sets of internals and different cone configurations. Data on differences in movement of filter material, air, and ash provided insight into the optimum GBF design.

Multicontaminant Control

Besides particulate removal, a GBF may be able to capture other coal contaminants through the use of chemically reactive filter media. Combustion Power Company (CPC) proposed to develop a reactive filter medium composed of limestone and clay for the removal of sulfur and alkali species, repectively. This approach was changed because of the well-evolved alternative methods of sulfur removal, the thermodynamic limitations of limestone for H_2S removal, and the problems associated with the oxidation of calcium sulfide to sulfate. CPC entered into a subcontract agreement with Research Triangle Institute (RTI) to investigate alternative reactive filter materials. The program is divided into four phases. In the first phase, RTI is investigating clay-based sorbents for control of alkali, primarily sodium vapors, under IGCC and PFBC conditions. Option I will focus on clay-based sorbents for the additional sorption of lead, cadmium, barium, beryllium, and chromium. Under Option II, the sodium-based sorbents will be investigated for the sorption of halogen and metalloid species, and Option III will involve investigation of Ni-based catalysts for ammonia decomposition.

GBF Testing at Power Systems Development Facility

SCS has entered into a cooperative agreement with DOE/Morgantown Energy Technology Center (METC) for the design, construction, and operation of a hot-gas cleanup test facility for gasification and pressurized combustion. This facility is being commissioned in Wilsonville, Alabama. Shakedown and testing of the GBF at SCS are scheduled for early 1997.

The basic filter design is patterned after successful test units developed at CPC in the 1970s and tested at NYU during the 1980s. Anticipated filter design changes necessary for scaleup for commercial testing are based on computational fluid dynamics simulations. Dimensions of the filter are adjustable. The filter diameter can be changed by altering the refractory lining to allow operation at different capacities. The depth of the filter can be changed from 10 to 5 ft, if it is determined that a shallower filtration bed is suitable for ash collection efficiency. System design of the GBF is the same as for a commercial unit.

Results/Accomplishments

GBF Component Development Program

In the cold flow tests in the component test facility (Wilson et al., 1995), basically all configurations yielded such similar characteristics that it was not possible to conclude that one configuration was superior. More elaborate, or longer-running, tests would be needed to yield conclusions.

A full-circumference, 3.5-ft-diameter filter with simulated refractory lining was built that closely duplicated the commercial arrangement. Difficulty with ash collection with the 6-mm filter material was encountered. This issue was not anticipated. As a result, testing concluded prematurely.

Findings from the cold-flow tests are as follows:

- Airflow measurements at the filter top surface were influenced by the surface topography due to the media angle of repose between the media fill pipes. The air velocity would consistently increase in the valleys. The lowest valley occurs at the wall, and the air velocity was highest at that location. When the surface was leveled, the top surface flow measurements were much more consistent.

- The media velocity across the top surface of the filter was constant except for lower velocity at the filter vessel wall. This velocity profile existed for all media and airflow combinations.

- When commercial operation near maximum filter capacity was simulated, an air bubble at the end of the filter inlet tube grew with the gas flow. When the airflow reached about 80 to 90 percent of minimum fluidization, the bubble bulged around the sides of the inlet and released, rising up about 12 in. before dissipating into the filter. This observation gives insight into the operation of the filter near maximum capacity.

- The filter pressure drop was examined as a function of superficial velocity. When the airflow reached minimum fluidization, the media began bubbling in regions of the filter. Instead of large bubbles forming at the top surface and splashing over the vessel side walls as the media started to fluidize, more material entered the vessel from the supply reservoir above the filter and the filter level continued rising.

Multicontaminant Control

The multicontaminant control work is being done at RTI. Some recent results on the
development of a clay-based filter material for alkali removal are reported here.

Sorbent Screening

Identification of Target Clay Materials. Emathlite, kaolin, bauxitic kaolin, attapulgite, and
calcium montmorillonite were selected as potential candidates for GBF media capable of alkali
removal. In addition to previous documentation as alkali getters, these materials were found to be
readily available in quantities sufficient for production of large amounts of GBF media in the
southeastern part of the United States. Alpha-alumina, a known sodium nongetter, was also added
to this list to provide a zero-reactivity baseline for alkali sorption. Loss on ignition (LOI),
chemical composition based on LOI, and bulk price per LOI weight for these materials are listed
in Table 1.

Alkali Sorption Screening Procedure. Problems generating and monitoring alkali vapor
complicate effective screening efforts. A simple screening procedure for alkali sorption was

Table 1. Chemical Composition and Bulk Cost of Various Clays[a]

	Emathlite 3000	Emathlite 600	Kaolin	Bauxitic Kaolin	IGB[b]	Attapulgite	Alumina
P_2O_5	3.48	3.75	0.30	0.09	0.12	0.99	0.00
SiO_2	68.97	68.79	51.15	35.77	53.78	61.76	0.00
Na_2O	0.3	0.40	0.10	0.06	1.47	0.07	0.00
K_2O	0.95	1.04	0.15	0.12	0.53	0.85	0.00
SO_3	0.14	0.18	0.12	0.26	1.45	0.30	0.00
MgO	2.95	2.73	0.10	0.04	5.32	10.38	0.00
CaO	5.43	5.83	0.10	0.06	12.59	4.81	0.00
Al_2O_3	8.33	7.75	44.17	57.59	17.68	10.41	100.00
TiO_2	0.38	0.39	1.73	2.41	0.10	0.46	0.00
Fe_2O_3	0.37	2.98	0.62	1.18	1.43	3.90	0.00
Loss on ignition (LOI)	9.81	8.87	13.78	23.94	20.87	21.61	0.00
Bulk cost ($/ton)	74.50	60.35	68.25	231.40	106.25	141.60	

[a] Mineral analysis and bulk cost reported as weight percent on LOI-free basis.
[b] Trade name for calcium montmorillonite.

developed by Dr. Schulz's group at the University of Surrey, U.K. (McLaughlin, 1990). Reagent-grade NaCl is added to the material being tested, producing a mixture with 10 wt% NaCl. Thermogravimetric analysis (TGA) records the weight loss of this sample as it is heated from room temperature to 1,000 °C. As the NaCl is heated, it begins to evaporate, releasing sodium vapor. An alkali nongetter will release the vapor products and experience a weight loss equivalent to the amount of salt in the sample. An alkali getter absorbs the sodium vapor and experiences a weight loss that is related to its alkali sorption activity.

Thermal Analysis. A DuPont 1090 thermal analysis system (Gupta and Gangwal, 1992) was used to screen the clay materials for alkali sorption activity in nitrogen, a simulated IGCC gas containing 87 vol% CO_2, 10 vol% H_2, and 3 vol% H_2O and a simulated PFBC gas with 6.9 vol% CO_2, 13 vol% O_2, 3 vol% H_2O, and the balance nitrogen. During these screening tests, a 20-mg sample was heated at 15 °C/min from room temperature to 1,000 °C in 50 mL/min of test gas.

The TGA screening results are summarized in Table 2 , which lists ultimate percent weight loss for all seven samples as a function of gas composition. As expected, α-alumina, a known non-getter, showed 10 percent weight loss for all three test gases, indicating no alkali sorption. Based on an average weight loss for the three test conditions, a list of the clay materials from most effective alkali getter to the known alkali nongetter is emathlite 600 > kaolin > emathlite 3000 > bauxitic kaolin > calcium montmorillonite (IGB) > attapulgite > α-alumina.

Sorption Activity for Various Heavy Metals. A series of TGA runs were performed to evaluate kaolin and emathlite 600 as potential high-temper-ature sorbents for Pb, Ba, Be, Cd, and Cr. Samples for these TGA runs were prepared by mixing 90 wt% of either kaolin or emathlite 600 and 10 wt% of a trace metal chloride. The samples were heated in the TGA at 15 °C/min to 1,000 °C in 50 mL/min of helium using the same procedure as described previously. The TGA results are summarized in Table 3, as final weight losses for both emathlite and kaolin

Table 2. TGA Reactivity of Various Clays

Clay Sample	Percent Weight Loss			
	N_2	IGCC	PFBC	Average
Emathlite 3000	8.40	5.65	6.09	6.71
Emathlite 600	5.75	5.50	6.23	5.83
Kaolin	7.04	5.37	7.03	6.48
Bauxitic kaolin	7.43	7.44	8.57	7.81
Calcium montmorillonite	8.60	7.86	7.68	8.05
Attapulgite	8.49	7.60	8.13	8.07
α-Alumina	10.24	10.39	10.32	10.32

Table 3. Summary of TGA Results

Trace Metal	Percent Weight Loss	
	Kaolin	Emathlite 600
$PbCl_2$	10.1	8.4
$CrCl_3$	9.8	7.2
$BaCl_2$	2.1	3.1
$BeCl_2$	4.5	6.1
$CdCl_2$	8.1	9.4

samples with each of the metal chlorides. Arranging the trace metals in lists of decreasing sorption capability produces Ba > Be > Cr > Pb > Cd for emathlite 600 and Ba > Be > Cd > Cr > Pb for kaolin.

GBF Media Development

Based on the TGA screening tests, emathlite and kaolin were identified as effective alkali sorbents. The next step was to identify a simple and inexpensive production process for converting the raw clays into GBF media. A GBF medium must possess mechanical strength and attrition resistance to survive the mechanical demands of a moving-bed process. For alkali removal, the GBF medium must also maintain a high level of reactivity with alkali vapor. Economic constraints for the preparation process require the ability to produce large quantities of material at minimal costs.

Figure 2 illustrates that high mechanical strength and chemical reactivity are a direct consequence of the binders, additive, and the pelletization method used. Identification of the correct combination for optimal strength and reactivity proceeded in iterative steps. Extrusion with spheronization and disk pelletization were selected as pelletization techniques based on economic and production capacity constraints. Pure emathlite and kaolin pellets were produced by each pelletization technique and tested for mechanical strength and reactivity. Binders and additives were used to improve pellet properties and achieve pellet design requirements.

Figure 3 shows crush strength and attrition loss for calcined kaolin and emathlite pellets prepared by both extrusion/spheronization and disk pelletization. From data reported in Figure 3, crush strength values for calcined kaolin and emathlite pellets produced by extrusion/ spheronization are almost an order of magnitude higher than those produced by disk pelletization. A similar trend is observed for attrition resistance. Because pellet production by disk pelletization is much less expensive than extrusion/spheronization, extensive binder and additive testing was performed in
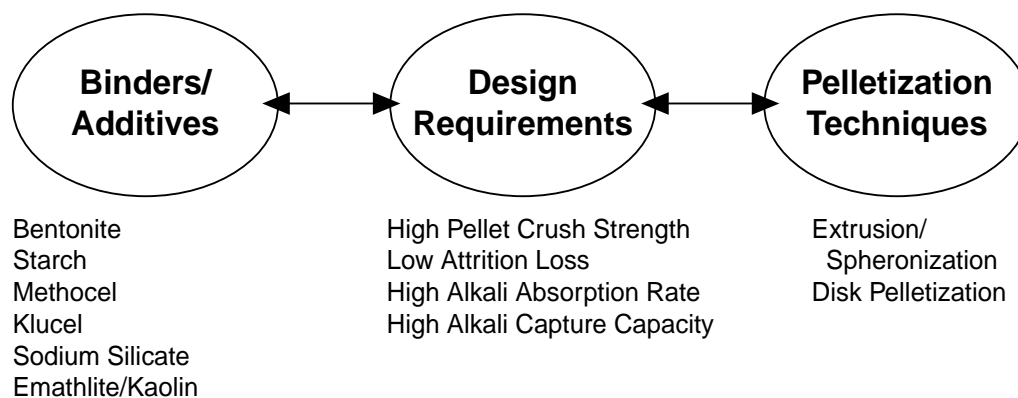

Binders/ Additives	Design Requirements	Pelletization Techniques
Bentonite	High Pellet Crush Strength	Extrusion/
Starch	Low Attrition Loss	Spheronization
Methocel	High Alkali Absorption Rate	Disk Pelletization
Klucel	High Alkali Capture Capacity	
Sodium Silicate		
Emathlite/Kaolin		

Figure 2. Approach to GBF Media Development

**Crush Strength
(ASTM D-4058)**

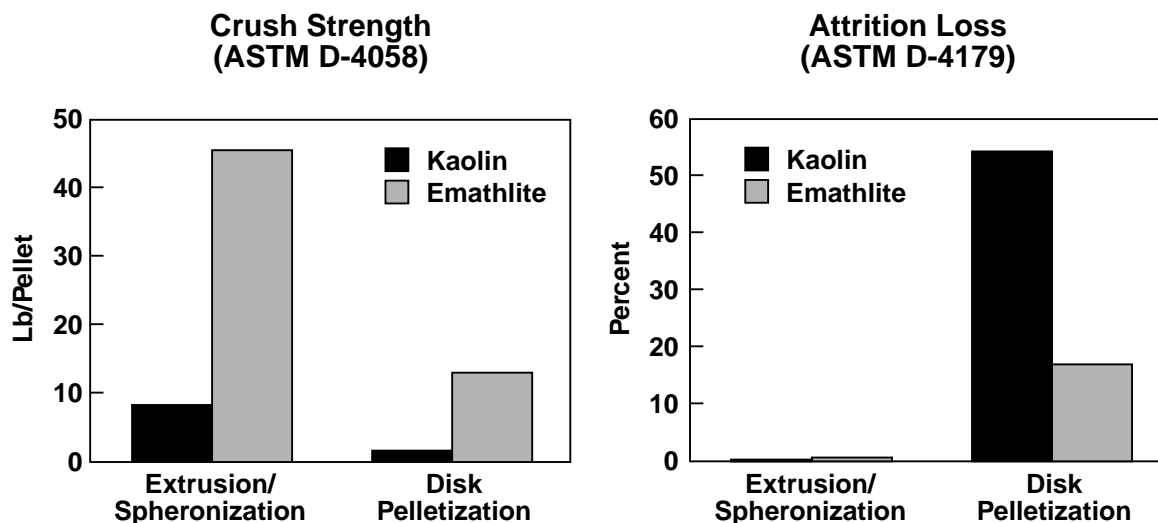**Attrition Loss
(ASTM D-4179)**

**Figure 3. Mechanical Properties of Kaolin at Emathlite Pellets
Produced By Extrusion and Disk Pelletization**

an attempt to improve the strength characteristics of pellets produced by disk pelletization. Unfortunately, the strength of pellets produced by disk pelletization could not be increased and testing of pellets produced by disk pelletization was discontinued.

Unlike disk pelletization, extrusion/spheronization produced strong attrition-resistant pellets with both kaolin and emathlite. Comparison of the strength characteristics of these extruded pellets indicates emathlite pellets possess significantly more strength than kaolin pellets. The TGA reactivity of these pellets, shown in Figure 4, clearly demonstrates kaolin pellets are more reactive. As binders and additives are more likely to affect mechanical strength than reactivity, research efforts were focused on increasing the mechanical strength of extruded kaolin pellets with binders and additives. As extruded emathlite pellets are very strong, emathlite was tested as a potential strengthening additive for extruded kaolin pellets.

Effects of binders and additives on mechanical strength of extruded kaolin pellets are summarized in Figure 5. A kaolin sorbent with 2 wt% sodium silicate binder (designated as KOS-1) emerged a winner in terms of crush strength and attrition resistance based on the data shown in Figure 5. Another promising candidate was a kaolin and emathlite mixture containing 35 wt% emathlite and 65 wt% kaolin (designated KE-35).

Alkali sorption activity for extruded pellets of kaolin (KO-2), emathlite (ET-1), kaolin with 2 wt% sodium silicate (KOS-1), and 35 wt% emathlite in kaolin (KE-35) is compared in Figure 6. From this figure, the weight increase associated with alkali absorption is 7.5×10^{-4} mg/min for KO-2, 3.7×10^{-4} mg/min for KOS-1, 3.0×10^{-4} mg/min for KE-35, and 2.3×10^{-4} mg/min for ET-1.
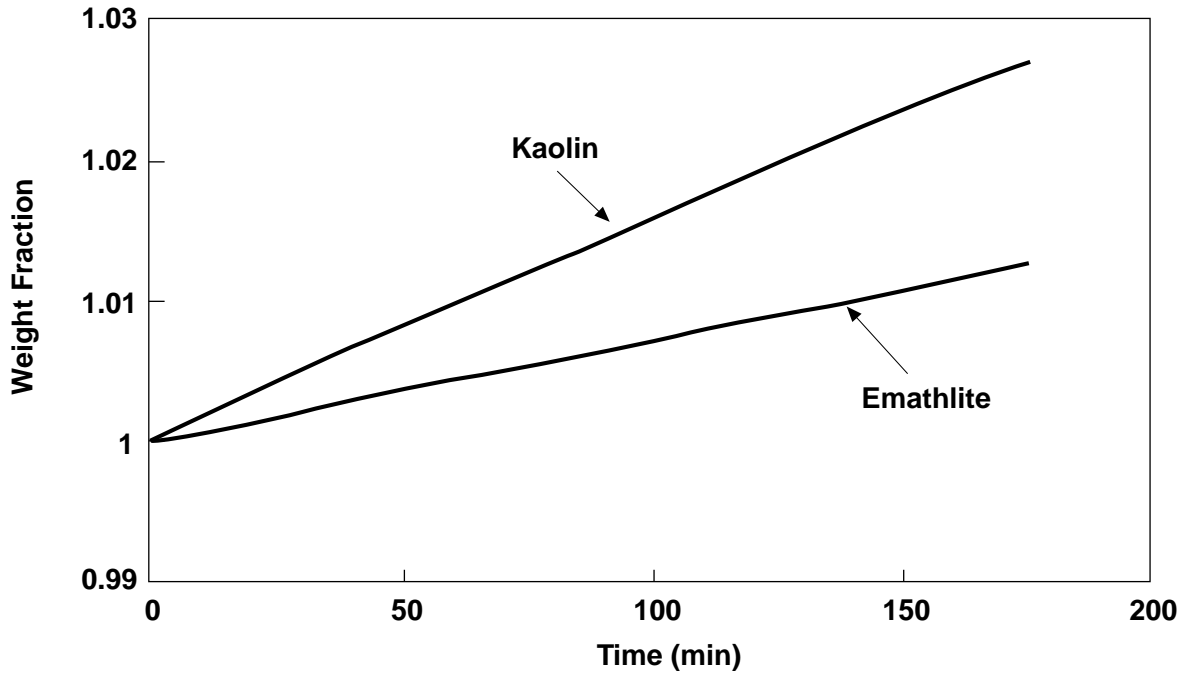
**Figure 4. Comparison to TGA Reactivity of Kaolin and Emathlite Extruded Pellets
With 40 ppm of Na Vapor at 870 °C**

Based on the chemical reactivity and mechanical strength measurements, KOS-1 was selected as the optimum formulation for bench-scale testing as well as lift-tube attrition testing. A 40-lb batch of this sorbent was prepared.
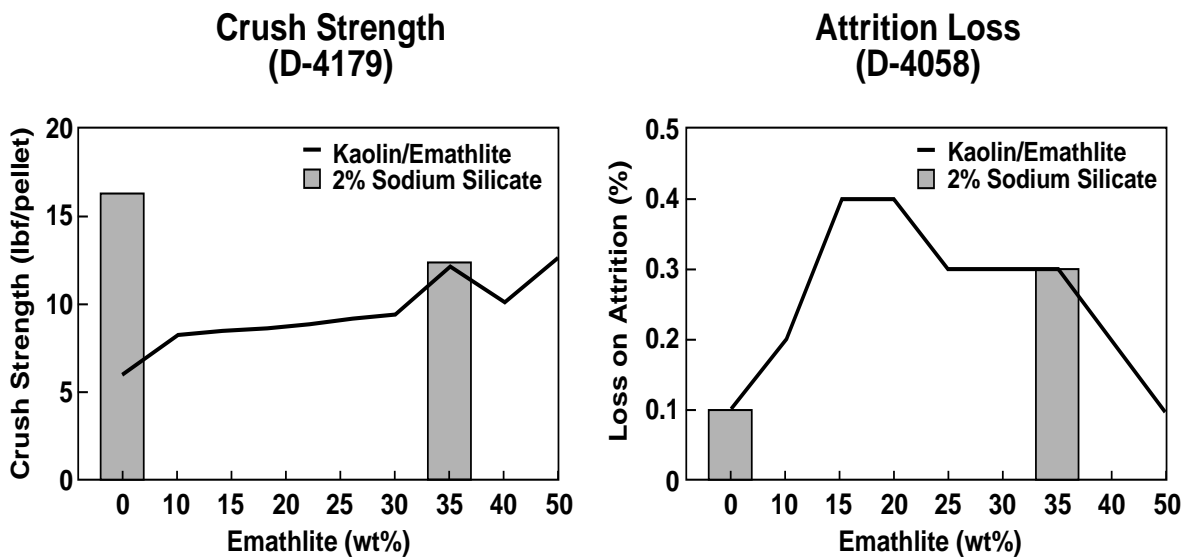

**Figure 5. Mechanical Strength of Kaolin Pellets
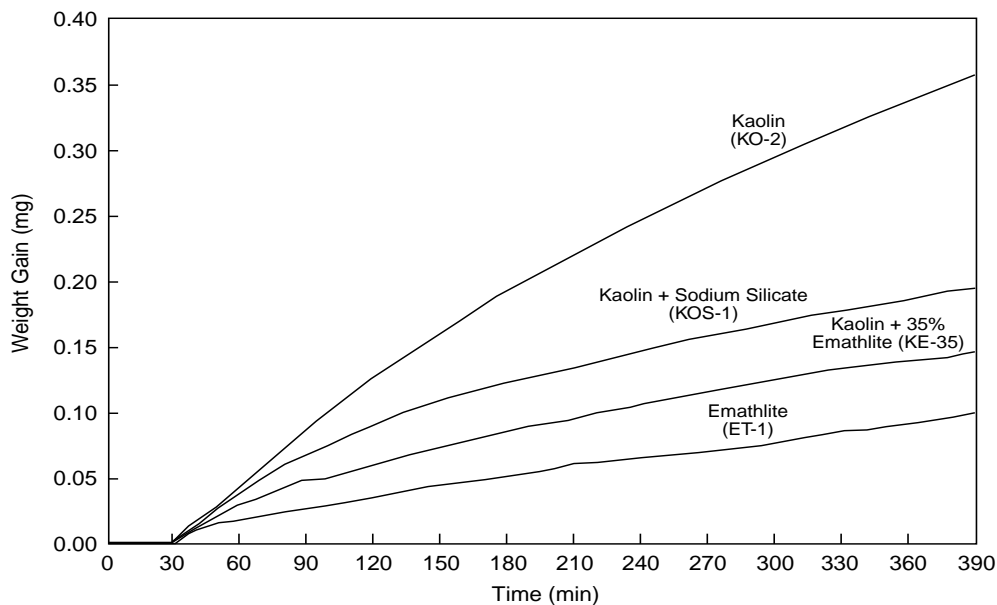as a Function of Emathlite Content**

**Figure 6. TGA Reactivity of Various Formulations At 870 °C
With 40 ppmv of Na Vapor in Nitrogen**

Bench-Scale Reactor Results. A schematic diagram of the bench-scale test system is shown in Figure 7. This reactor system is composed of a gas generation system, preheating section, and a quartz reactor. The gas delivery system can simulate a gas stream generated from coal combustion in either a PFBC or an IGCC system. Steam is generated from a metered water flow, which is mixed with a gas stream in the preheater section. The gas generation system can also provide a stream containing HCl to achieve a concentration of 100 ppmv in gas entering the reactor. This HCl stream is mixed into the gas after the preheating section to minimize system corrosion resulting from an HCl and steam mixture. Rather than having one main gas supply stream, the main gas supply has been split into two separate streams to generate 40 ppmv of sodium vapor in the reactor by salt vaporization.

The preheater section serves to vaporize the water generating steam, to reduce heating demands in the furnace, and to maximize salt vaporization during sodium vapor generation.

Problems with sodium vapor corrosion and condensation were minimized by generating the sodium vapor in the reactor near the sorbent bed. Sodium vapor is generated by passing a fraction of the gas supply through a bed of approximately 10 to 15 g of coarse salt crystals between 700 and 800 °C. The amount of sodium vaporized is controlled by the fraction of gas supply forced through the salt bed and the temperature of the salt bed. Furnace heating zones and set points have been optimized to minimize the temperature gradient in the sorbent bed. The temperature gradient in the top half of the bed, where a majority of the reaction is expected to
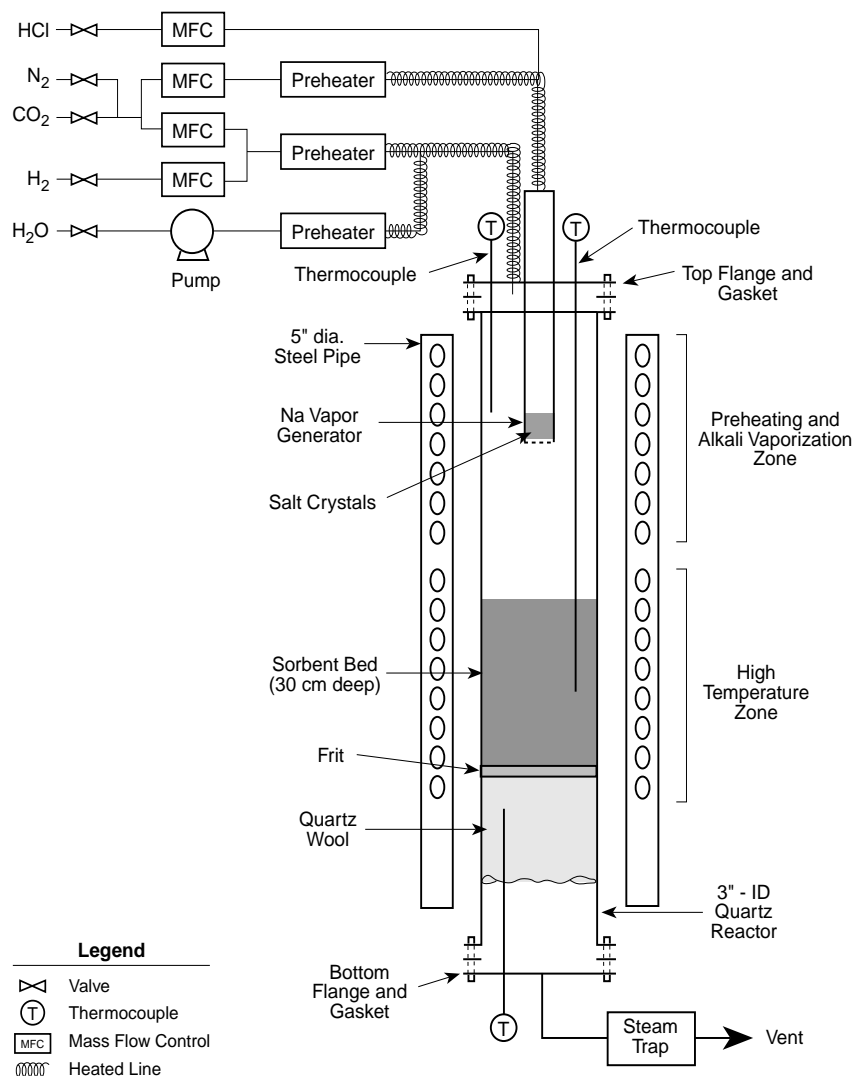
HCl

MFC

N₂

MFC Preheater

CO₂

MFC

H₂

MFC Preheater

H₂O

Pump Preheater

Thermocouple

Thermocouple

Thermocouple

Top Flange and Gasket

5" dia. Steel Pipe

Na Vapor Generator

Salt Crystals

Preheating and Alkali Vaporization Zone

Sorbent Bed (30 cm deep)

High Temperature Zone

Frit

Quartz Wool

3" - ID Quartz Reactor

Legend

⋈ Valve

(T) Thermocouple

MFC Mass Flow Control

⌒⌒⌒ Heated Line

Bottom Flange and Gasket

Steam Trap → Vent

Figure 7. Schematic Diagram of Bench Unit

take place, is less than 5 °C. Any sodium vapor managing to escape the sorbent bed is trapped in a cold section of the reactor filled with quartz wool. Effluent from the reactor is vented through a steam trap to collect any condensed steam.

Because removing sequential layers in a precise and systematic fashion is important to modeling the alkali absorption profile in the sorbent bed, a specialized vacuum sampling system was designed and constructed. This sampling system has just enough vacuum to remove and collect a known depth of sorbent. Successive layers of the sorbent bed are removed by lowering the sampling system.

Sorbent tests of 150 hours of continuous operation are scheduled to begin with the KOS-1 sorbent in the near future. Results from these tests will be used to develop a kinetic model for alkali absorption of the most promising sorbent formulation.

GBF Testing at Power Systems Development Facility

The filter is currently under construction in Wilsonville, Alabama. Checkout and shakedown are scheduled in the first quarter of 1997 followed by combustion and gasification tests. The test objectives are to establish a filter material size suitable for particulate collection, determine filter capacity, and optimize filter efficiency.

Application

It is technically advantageous to capture coal-derived contaminants as well as particulate matter in a single vessel. The gas stream cleanup process in advanced power system would be simpler with a single vessel, and capital and operating costs would be lower. A GBF is a perfect candidate for multicontaminant control. The test program to develop chemically reactive filter material is showing good results with alkali control utilizing kaolin and emathlite clays.

Future Activities

Other options for chemically reactive filter material include a sodium-based filter material for the control of halogens and metalloids in reducing gas environments, and a nickel catalyst for the destruction of ammonia and hydrogen cyanide in a reducing gas environment. A single filter that can accomplish control of particulate and one or more of these other contaminants has an advantage in advanced power plant applications.

Combustion and gasification tests are planned at the PSDF to determine filter capacity, optimize filter efficiency, and compare different filter materials. These tests are scheduled to begin during the first quarter of 1997. Testing is scheduled to continue until late 1999. Filter material developed for contaminant control could be made available for testing at the PSDF.

Acknowledgements

The METC Contracting Officer's Representative (COR) is Mr. Thomas P. Dorchak. The period of performance is from October 1992 to November 1996 for the work being done for METC. Multicontaminant control filter material is being developed under subcontract to RTI. The testing at SCS is scheduled from 1997 to 1999.

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